

DIRECT NUCLEAR DEMAGNETIZATION OF HIGH DENSITY
BODY-CENTERED-CUBIC (BCC) AND HEXAGONAL-CLOSE-PACKED (HCP)
SOLID ^3He

By

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To my family.

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Direct nuclear demagnetization was used to cool high-density solid ^3He into the magnetically ordered phases. The magnetic state of the ^3He during precooling and demagnetization was indicated by pressure change detected by a capacitive strain gauge. The entropy of the samples was reduced by precooling to temperatures between 0.55 and 3.99 mK in a field of 2.5 T. For a bcc sample with a molar volume of $20.10 \text{ cm}^3/\text{mole}$ and remaining entropy of approximately $0.01 R \ln 2$ two magnetic transitions were observed. A similar experiment on two hcp samples with molar volumes of $19.65 \text{ cm}^3/\text{mole}$ and $19.65 \text{ cm}^3/\text{mole}$ was carried out. Pressure data at various entropies indicated different behaviors near zero field: a decrease in pressure occurred for low-entropy samples, with an increase in pressure

for high-entropy samples. Results from the high-temperature expansion of the multiple-exchange and Heisenberg models can explain only the pressure data with remaining entropies $0.72R\ln 2$ or higher. We conclude that the pressure behavior observed in samples with entropies of 0.46, 0.23 and $0.02R\ln 2$ are the results of magnetic ordering in hcp solid ^3He .

CHAPTER 1 INTRODUCTION

Solid ^3He becomes magnetically ordered at temperatures ≤ 1 mK via exchange interactions made possible because of its large zero point motion. Despite its simple structure, ^3He has rich magnetic properties. The body-centered-cubic (bcc) solid in a magnetic field has three magnetic phases: the paramagnetic phase (PP), the low field phase (LFP), and the high field phase (HFP).

While most of the magnetic properties of the bcc solid are well characterized, there exists little information about the properties of the hexagonal-close-packed (hcp) solid which forms at high pressure. Theoretical calculations [1] and experimental works [2, 3, 4] indicate that the solid would order ferromagnetically near $20\text{ }\mu\text{K}$. Such a low temperature makes the study of the magnetic properties in the hcp solid difficult. Despite many attempts [2, 3, 4], no magnetic transition in the hcp solid was observed because the ordering temperature could not be reached using the indirect cooling method.

To achieve lower temperatures, ^3He samples may be directly demagnetized, an idea that has been around for many years [5]. This method was recently employed at the University of Tokyo [6] on both bcc and hcp solids. Their results showed that the bcc solid reached both HFP and LFP implying, that the sample was cooled below $28\text{ }\mu\text{K}$ (extrapolated from susceptibility data of high density bcc solid ^3He [7]). For their two hcp samples, no clear signature of the ordering was observed because of heating of the cell near the end of the demagnetization.

Using the sample demagnetization technique, we carried out pressure measurements for one bcc and two hcp samples. The pressure changes of the samples during the precooling and demagnetization indicated the magnetic state of the sample. With an improved design of the experimental cell, we have reduced unnecessary heating near the end of the sample demagnetization and thus eliminated the problem encountered by the Tokyo group. Also, we have made the first clear observation of the pressure signature of magnetic ordering in the hcp solid ^3He .

This thesis is arranged into five chapters. In chapter one, a review is given of the current understanding of the nuclear magnetic properties of solid ^3He . There will be a discussion of the existing theoretical models and a summary of previous experiments on solid ^3He . Chapter two is devoted to the experimental arrangements used in this work. A detailed description of the experimental apparatus and data-taking process will be provided. Chapter three is about the experimental procedures such as calibration of the strain gauges and temperature regulation. The experimental results and their contribution toward our understanding of nuclear magnetism in solid ^3He are presented in chapter four together with the simulation data of sample demagnetization using the multiple exchange and Heisenberg models. Chapter five is a summary of this experiment with a proposal for future works.

1.1 Pressure and Magnetic Phase Diagrams of Solid ^3He

Because of weak nuclear interactions, magnetic ordering of nuclear spins in most solids exists only in the nanokelvin temperature range or lower. In ^3He , however, ordering takes place at a few mK for some density and magnetic field. Such a high ordering temperature is brought about by particle exchange made possible because ^3He has a large zero-point motion. The wave function of one atom overlaps with those of its neighbors leading to the

exchange of their positions in the lattice. Based on nuclear magnetic resonance (NMR) data [8], the exchange rate in ^3He is estimated to be as high as 20 MHz at melting pressure.

The pressure versus temperature ($P - T$) phase diagram of ^3He is shown in Fig. 1.1. As a result of the large zero-point motion, ^3He remains liquid at low pressure even at $T = 0$ and does not solidify until its pressure reaches almost 3 MPa. Between 3 and 10 MPa, solid ^3He has a body-centered-cubic (bcc) structure, again as a result of the large zero-point motion. At melting pressure, the solid orders into an antiferromagnetic phase with a Néel temperature T_N of 0.934 mK [9] at zero magnetic field. As the solid is compressed at high pressure, it is more difficult for the atoms to exchange their places because of the hard-core potential. The difficulty of the atoms to exchange at high density is analogous to the difficulty of moving around in a crowded bus; the denser the crowd, the harder it is for two passengers to exchange their positions [10]. The exchange frequency becomes smaller at high pressure,¹ causing a rapid decrease in the ordering temperature, which drops below 40 μK near 10 MPa at zero field. There is a simple empirical equation relating the ordering temperature to the molar volume of a bcc sample [11]:

$$T_N = v^{16.5 \pm 1}, \quad (1.1)$$

where, v the molar volume of the solid, ranges from 24.14 to 22.46 cm^3/mole , corresponding to the solid pressures from 3.52 to 5.32 MPa respectively.

Above 10 MPa, the crystal structure of solid ^3He is hexagonal-close-packed (hcp). Because of the lattice symmetry which favors three-particle exchange, the hcp solid is expected to order into a ferromagnetic phase [1]. Previous experimental results also indicate a strong

¹The exchange frequency also decreases for processes which involve more than four particles due to a longer path length.

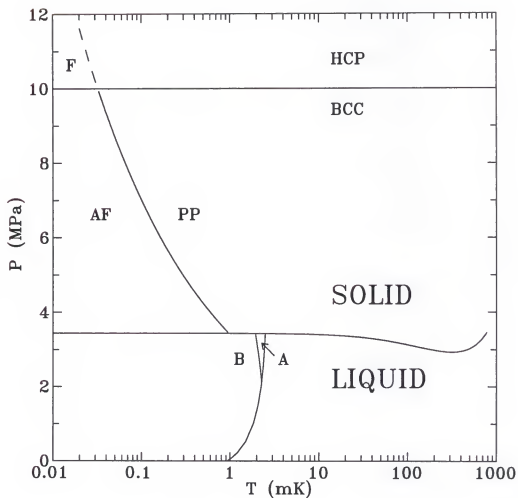


Figure 1.1. P-T phase diagram of ^3He at zero magnetic field where F, AF and PP indicate the ferromagnetic, anti-ferromagnetic, and paramagnetic phases of the solid respectively. A and B show the superfluid phases of the liquid.

ferromagnetic tendency [2, 3, 4]. An hcp sample near the bcc-hcp boundary line is expected to order at about 20 μK [3]. Such a low ordering temperature makes the study of the magnetic properties in the hcp solid difficult.

Despite many attempts to reach the ordered phase either by the indirect [2, 3, 4] or direct [6] cooling methods, no clear evidence of magnetic ordering in the hcp solid has been observed. The main goal of our work, therefore, is to cool an hcp sample into its ordered state, identify the ordering signature, and characterize the ordered state.

Unlike the hcp solid, the magnetic properties of the bcc solid are well characterized, particularly at melting density ($v_{\text{melt}} = 24.22 \text{ cm}^3/\text{mole}$). There are many advantages to studying a solid sample on the melting curve. With the presence of the liquid, the solid density is more uniform. The high ordering temperature (about 1 mK) can be easily reached by nuclear demagnetization refrigerators. Excellent thermal contact between the solid and the refrigerator can be provided by the liquid. A low pressure, about 3 MPa compared to 10 MPa of a hcp sample, yields higher sensitivity in pressure measurements because the diaphragm of the pressure gauge can be made thinner.² Experiments on higher-density bcc solids [11, 12] show that many of the magnetic properties can be scaled to those at the melting density using a simple relation such as Eq. 1.1.

Despite its simple lattice structure, ^3He has surprisingly rich magnetic properties. Figure 1.2, taken from Ref. [13], shows the magnetic-field-versus-temperature ($B - T$) phase diagram of solid ^3He at its melting density. There exist three magnetic phases: the paramagnetic phase (PP), the low-field phase (LFP), and the high-field phase (HFP). In the LFP, the nuclear spins have an up-two-down-two (U2D2) structure [14]. It is made up of

²The pressure change in the sample is detected by the deflection of a metal diaphragm. A thin diaphragm deflects more than a thick one at a given pressure. See section 2.1 for discussion on the construction of the pressure gauge.

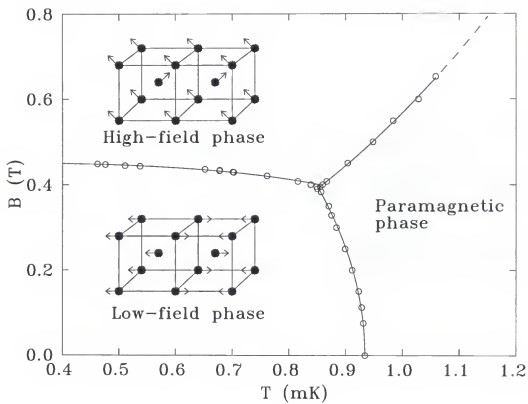


Figure 1.2. B-T phase diagram of bcc solid ^3He at melting pressure.

ferromagnetic planes normal to one of the three principal cubic axes, with spins pointing in one direction in two planes and pointing in the opposite direction in the next two planes. This periodic alternation of spin direction continues throughout the entire magnetic domain of the crystal. When the applied field is larger than the critical field, B_{c1} , the solid enters the high-field phase. At melting density, B_{c1} is about 0.45 Tesla and decreases rapidly with increasing solid density. In 1991, Fukuyama and co-workers [12] measured B_{c1} for molar volumes ranging from 24.06 to 22.45 cm³/mole and found that at zero temperature, $B_{c1}(0) = v^{14.5 \pm 0.3}$. In the HFP, the spins are believed to order in a canted antiferromagnetic structure which is composed of two simple-cubic sublattices with spin directions tilted away from the direction of the applied field. The angle ϕ between the spins and the field is equal in both sublattices. It decreases with increasing field and eventually becomes zero at the HFP-PP transition at the second critical field $B_{c2}(0)$ at zero temperature. At melting density, $B_{c2}(0)$ is expected to be about 22 Tesla [15]. The existence of two magnetically ordered phases in the bcc solid is believed to result from the competition between different exchange processes. A detailed explanation can be found in the next section.

1.2 Review of Exchange Theory of ³He Magnetism

Currently, there is no existing theoretical work, including the well-known multi-particle exchange theory [1, 16], that can accurately describe all the magnetic properties of solid ³He. In any case, a closer look at this theory is worthwhile, since it is the only model which can explain most of the complicated characteristics of magnetism in solid ³He. A simple model for describing exchange of particles is the Heisenberg nearest-neighbor model (HNN) which takes into account only the pairwise exchanges of an atom and its nearest neighbors.³ It can be easily shown that a two-particle exchange process causes ³He to order

³In the hcp solid, three-particle exchange processes also lead to the HNN model.

antiferromagnetically at low temperature. The proof and a discussion of the HNN model is given below.

1.2.1 Heisenberg Nearest-Neighbor Model

^3He atoms are fermions with spin $1/2$. The Pauli principle states that the wave function of two fermions must be antisymmetric when the particles are interchanged. In other words, two identical fermions cannot occupy the same quantum state. Mathematically, this principle can be expressed as:

$$\psi_{12} = -\psi_{21} \quad (1.2)$$

where, ψ_{12} is the total wave function of the two fermions. To check the validity of this equation, one can make the assumption that the two fermions can occupy the same quantum state. Equation 1.2 becomes $\psi_{12} = -\psi_{12} = 0$, *i.e.* the amplitude of the wave function is zero.

The wave function, ψ_{12} , can be broken down into two parts:

$$\psi_{12} = \phi_{12}\chi_{12}, \quad (1.3)$$

where ϕ_{12} is the spatial wave function and χ_{12} the spin state of the two fermions. For the ground state, ϕ_{12} must be symmetric; that is $\phi_{12} = +\phi_{21}$. For two spin- $1/2$ particles, the total spin function χ_{12} can be either: $|\uparrow_1\uparrow_2\rangle$, $\frac{1}{\sqrt{2}}(|\uparrow_1\downarrow_2\rangle + |\downarrow_1\uparrow_2\rangle)$, $\frac{1}{\sqrt{2}}(|\uparrow_1\downarrow_2\rangle - |\downarrow_1\uparrow_2\rangle)$, or $|\downarrow_1\downarrow_2\rangle$. Since ϕ_{12} is symmetric and the total wave function $\psi_{12} = \phi_{12}\chi_{12}$ must be antisymmetric, the Pauli principle requires that χ_{12} be antisymmetric. Out of the four choices only $\frac{1}{\sqrt{2}}(|\uparrow_1\downarrow_2\rangle - |\downarrow_1\uparrow_2\rangle)$ has the correct symmetry. This spin function yields a total spin of zero which implies antiferromagnetism. It can be shown that for an even number

of particle exchange, such as 2, 4, ..., the resulting ordered state is antiferromagnetic. For odd number of particle exchange, the process favors a ferromagnetic state (see articles by Roger *et al.* [1] and Thouless [16] for more detail). It is believed that the competition between these antiferromagnetic (even) and ferromagnetic (odd) processes brings about the existence of the LFP.

The Hamiltonian of solid ^3He is straightforward,

$$H = H_{Phonon} + H_{Exchange} + H_{Zeeman} + H_{Dipole}. \quad (1.4)$$

The energy scale for each term is: $H_{Phonon} \approx 10$ K, $H_{Exchange} \approx 1$ mK (at melting pressure), $H_{Zeeman} \approx 0.7$ mK (at 1 Tesla), and $H_{Dipole} \approx 100$ nK. Because of the large separation between these values, only the exchange and Zeeman terms dominate the thermodynamic properties of the solid for the temperature range between $10 \mu\text{K}$ to 100 mK. Equation 1.4 can be reduced to

$$H = H_{Exchange} + H_{Zeeman}, \quad (1.5)$$

and the partition function can be written as

$$Z = \text{trace} \{e^{-\beta H}\} = \text{trace} \{e^{-\beta(H_{Exchange} + H_{Zeeman})}\}, \quad (1.6)$$

where $\beta \equiv 1/k_B T$ with k_B the Boltzmann constant. For the exchange of two nearest neighbors, the Hamiltonian has the form

$$H_{Exchange} = -2J_{nn} \sum_{i < j} \vec{S}_i \cdot \vec{S}_j, \quad (1.7)$$

which commutes with the Zeeman Hamiltonian,

$$H_{Zeeman} = -\gamma\hbar/2 \sum_i \vec{S}_i \cdot \vec{B}_0, \quad (1.8)$$

where $\vec{S}_i$ is the spin operator in units of $\hbar$ at the i th lattice site, γ is the gyromagnetic ratio with a value of $\gamma/2\pi = 32.43$ MHz/Tesla, and $\vec{B}_0$ is the applied field. Therefore Eq. 1.6 becomes

$$Z = \text{trace}\{e^{-\beta(H_{Exchange})}e^{-\beta(H_{Zeeman})}\}. \quad (1.9)$$

To study the effect of the exchange interaction in ^3He , one need not cool the sample into its ordered state. Even at high temperature ($T \gg |J_{nn}|/k_B$), the impact of the interaction on the solid is large and can be measured through different thermodynamics quantities such as pressure, heat capacity, and susceptibility. Mathematically, the effect at high temperature can be shown by first writing down the relation between the free energy and the partition function,

$$F = -\frac{1}{\beta} \ln Z. \quad (1.10)$$

Since the temperature is assumed to be high, the exponential terms in Z can be expanded in powers of β . Equation 1.10 then becomes

$$F = -\frac{N_A}{\beta} \left[\ln 2 + e_2 \frac{\beta^2}{8} - e_3 \frac{\beta^3}{24} + \cdots + \frac{1}{8} (\gamma\hbar B_0 \beta)^2 (1 + k_B \theta_w \beta + \frac{\alpha_2 \beta^2}{8} + \cdots) + \cdots \right], \quad (1.11)$$

where N_A is Avogadro's number and e_2, e_3 and θ_w etc. are related to the exchange energy. For the Heisenberg nearest-neighbor model, $e_2 = 12J_{nn}^2$, $e_3 = 12J_{nn}^3$, and $\theta_w = xJ_{nn}/2k_B$, where x = number of nearest neighbors, which is 8 for the bcc and 12 for the hcp solid. To

obtain the value of pressure of the solid, one takes the negative of the derivative of F with respect to the volume, giving

$$P = N_A \left(\frac{\beta}{8} \frac{\partial \epsilon_2}{\partial v} - \frac{\beta^2}{24} \frac{\partial \epsilon_3}{\partial v} + \dots \right) + \left(\frac{N_A \gamma \hbar \beta B_0}{2} \right)^2 \left(\frac{k_B}{2} \frac{\partial \theta_w}{\partial v} + \frac{\beta}{2} \frac{\partial \alpha_2}{\partial v} + \dots \right). \quad (1.12)$$

The heat capacity can be calculated by taking the second derivative of the free energy with respect to the temperature. In zero field and at high temperature, one finds

$$C_v = -T \frac{\partial^2 F}{\partial T^2} = \frac{N_A k_B}{4} (\epsilon_2 \beta^2 - \epsilon_3 \beta^3 + \dots). \quad (1.13)$$

The susceptibility, given by the second derivative of the free energy with respect to the magnetic field, is

$$\chi = -\frac{\mu_0}{v} \left(\frac{\partial^2 F}{\partial B_0^2} \right) = \frac{C}{T - \theta_w + \frac{B}{T}}, \quad (1.14)$$

where $B = \theta_w^2 - \alpha_2/8k_B^2$, and $C = \mu_0 N_A (\gamma \hbar/2)^2 / k_B v$ with μ_0 the permeability of free space and v the molar volume. Using the HNN model gives $\theta_w = 4J_{nn}/k_B$ and $B = 4(J_{nn}/k_B)^2$ for a bcc solid.

1.2.2 Multiple-Particle Exchange Model

The simple pairwise exchange interaction of the HNN model alone cannot explain many of the experimental results. For example, the model predicts the ordered state of a bcc solid to be a normal antiferromagnet in which the atoms at the vertices have their spins up while the center atoms have their spins down. However, NMR [14] and neutron scattering results [17] indicate that the ground state actually has an up-two-down-two configuration. Another example of the insufficiency of the model is that the ordering transition is predicted to be second order and to occur at 2 mK. However, in 1974 Halperin *et al.* [18] found the

transition to be of first order, occurring at about 1 mK. In 1975, McMahan and Wilkins [19] pointed out the possibility that two-particle exchange might occur less frequently than other exchange processes, their reasoning being that the steric effect for two particle exchange is larger than other exchanges. Using the crowded bus analogy mentioned earlier, one can see that it requires more room for two passengers on the bus to exchange their places while less room is needed if three or four persons agree to switch their places in a cyclic fashion.

The general form of the multi-particle-exchange Hamiltonian is [1, 16]

$$H_{exchange} = \sum_{n,\alpha} J_{n\alpha} (-1)^P P_n, \quad (1.15)$$

where n is the number of fermions involved in an exchange process and α represents a specific process in the n -particle exchange. For example, n equals two for a nearest-neighbor exchange process. The two-particle process also includes the second, third-nearest-neighbor exchanges which have different exchange frequencies; α is therefore needed to distinguish each of these cases. $(-1)^P$ is 1(-1) for permutation involving an even (odd) number of particles. P_n is the permutation operator of n particles and $J_{n\alpha}$ is the exchange frequency for each permutation.

For an exchange process involving more than two particles ($n > 2$), both directions (clock-wise and counter-clock-wise) of the process must be taken into account. The exchange Hamiltonian becomes

$$H_{exchange} = \sum_{n,\alpha} J_{n\alpha} (-1)^P \mathcal{P}'_n, \quad (1.16)$$

where $\mathcal{P}'_2 = P_2$ and $\mathcal{P}'_n = (P_n + P_n^{-1})$ for $n > 2$. The different directions are represented by P_n and P_n^{-1} . The permutation operator P_n can be expressed in terms of spin operators.

Godfrin and Osheroff [20] give these expressions for exchange interactions of two, three, four, five and six particles. For two-particle exchange, $\mathcal{P}'_2$ has the form

$$\mathcal{P}'_2 = P_{ij} = \frac{1}{2}(1 + \sigma_i \cdot \sigma_j) = \frac{1}{2} + 2\vec{S}_i \cdot \vec{S}_j, \quad (1.17)$$

where the Pauli spin matrices σ_i are replaced by the spin-1/2 operators $\vec{S}_i$. For the three-spin exchange,

$$\mathcal{P}'_3 = P_{ijk} + P_{ijk}^{-1} = \frac{1}{2} + 2(\vec{S}_i \cdot \vec{S}_j + \vec{S}_j \cdot \vec{S}_k + \vec{S}_k \cdot \vec{S}_i). \quad (1.18)$$

Note that P_{ijk} is just another way to write P_3 . For an exchange process involving four particles,

$$\mathcal{P}'_4 = P_{ijkl} + P_{ijkl}^{-1} = \frac{1}{4} + \sum_{\mu < \nu} \vec{S}_\mu \cdot \vec{S}_\nu + 4G_{ijkl}, \quad (1.19)$$

where the summation runs over the six distinct pairs (μ, ν) of the four spins (i, j, k, l) and

$$G_{ijkl} = (\vec{S}_i \cdot \vec{S}_j)(\vec{S}_k \cdot \vec{S}_l) + (\vec{S}_j \cdot \vec{S}_k)(\vec{S}_l \cdot \vec{S}_i) - (\vec{S}_i \cdot \vec{S}_k)(\vec{S}_j \cdot \vec{S}_l). \quad (1.20)$$

If one explicitly writes out every term of the summation then

$$\begin{aligned} P_{ijkl} + P_{ijkl}^{-1} &= \frac{1}{4} + \vec{S}_i \cdot \vec{S}_j + \vec{S}_j \cdot \vec{S}_k + \vec{S}_k \cdot \vec{S}_l + \vec{S}_l \cdot \vec{S}_i + \vec{S}_i \cdot \vec{S}_k + \vec{S}_j \cdot \vec{S}_l + \\ &4[(\vec{S}_i \cdot \vec{S}_j)(\vec{S}_k \cdot \vec{S}_l) + (\vec{S}_j \cdot \vec{S}_k)(\vec{S}_l \cdot \vec{S}_i) - (\vec{S}_i \cdot \vec{S}_k)(\vec{S}_j \cdot \vec{S}_l)]. \end{aligned} \quad (1.21)$$

For five-spin exchange,

$$\mathcal{P}'_5 = P_{ijklm} + P_{ijklm}^{-1} = \frac{1}{8} + \frac{1}{2} \sum_{\mu < \nu} \vec{S}_\mu \cdot \vec{S}_\nu + 2 \sum_{\mu < \nu < \eta < \epsilon} G_{\mu\nu\eta\epsilon}. \quad (1.22)$$

Finally, for the exchange of six spins

$$\mathcal{P}'_6 = P_{ijklmn} + P_{ijklmn}^{-1} = \frac{1}{16} + \frac{1}{4} \sum_{\mu < \nu} \vec{S}_\mu \cdot \vec{S}_\nu + \sum_{\mu < \nu < \eta < \epsilon} G_{\mu\nu\eta\epsilon} + 4S_{ijklmn}, \quad (1.23)$$

where

$$\begin{aligned} S_{ijklmn} = & [(\vec{S}_i \cdot \vec{S}_j)(\vec{S}_k \cdot \vec{S}_l)(\vec{S}_m \cdot \vec{S}_n) + (\vec{S}_j \cdot \vec{S}_k)(\vec{S}_l \cdot \vec{S}_m)(\vec{S}_n \cdot \vec{S}_i)] \\ & - [(\vec{S}_i \cdot \vec{S}_j)(\vec{S}_k \cdot \vec{S}_m)(\vec{S}_l \cdot \vec{S}_n) + (\vec{S}_j \cdot \vec{S}_k)(\vec{S}_l \cdot \vec{S}_n)(\vec{S}_m \cdot \vec{S}_i) + (\vec{S}_k \cdot \vec{S}_l)(\vec{S}_m \cdot \vec{S}_i)(\vec{S}_n \cdot \vec{S}_j) \\ & + (\vec{S}_l \cdot \vec{S}_m)(\vec{S}_n \cdot \vec{S}_j)(\vec{S}_i \cdot \vec{S}_k) + (\vec{S}_m \cdot \vec{S}_n)(\vec{S}_i \cdot \vec{S}_k)(\vec{S}_j \cdot \vec{S}_l) + (\vec{S}_n \cdot \vec{S}_i)(\vec{S}_j \cdot \vec{S}_l)(\vec{S}_k \cdot \vec{S}_m)] \\ & + [(\vec{S}_i \cdot \vec{S}_l)(\vec{S}_j \cdot \vec{S}_n)(\vec{S}_k \cdot \vec{S}_m) + (\vec{S}_j \cdot \vec{S}_m)(\vec{S}_k \cdot \vec{S}_i)(\vec{S}_l \cdot \vec{S}_n) + (\vec{S}_k \cdot \vec{S}_n)(\vec{S}_l \cdot \vec{S}_j)(\vec{S}_m \cdot \vec{S}_i)] \\ & + [(\vec{S}_i \cdot \vec{S}_j)(\vec{S}_k \cdot \vec{S}_n)(\vec{S}_l \cdot \vec{S}_m) + (\vec{S}_j \cdot \vec{S}_k)(\vec{S}_l \cdot \vec{S}_i)(\vec{S}_m \cdot \vec{S}_n) + (\vec{S}_k \cdot \vec{S}_l)(\vec{S}_m \cdot \vec{S}_j)(\vec{S}_n \cdot \vec{S}_i)] \\ & - [(\vec{S}_i \cdot \vec{S}_l)(\vec{S}_j \cdot \vec{S}_m)(\vec{S}_k \cdot \vec{S}_n)]. \end{aligned} \quad (1.24)$$

Substituting all of the listed cycles back into Eq. 1.16, one gets

$$\begin{aligned} H_{exchange} = & \sum_{\alpha} \left[- \sum_{i < j} J_{2\alpha} \left(\frac{1}{2} + 2\vec{S}_i \cdot \vec{S}_j \right) \right. \\ & + \sum_{i < j < k} J_{3\alpha} \left(\frac{1}{2} + 2(\vec{S}_i \cdot \vec{S}_j + \vec{S}_j \cdot \vec{S}_k + \vec{S}_k \cdot \vec{S}_i) \right) \\ & - \sum_{i < j < k < l} J_{4\alpha} \left(\frac{1}{4} + \sum_{\mu < \nu} \vec{S}_\mu \cdot \vec{S}_\nu + 4G_{ijkl} \right) \\ & + \sum_{i < j < k < l < m} J_{5\alpha} \left(\frac{1}{8} + \frac{1}{2} \sum_{\mu < \nu} \vec{S}_\mu \cdot \vec{S}_\nu + 2 \sum_{\mu < \nu < \eta < \epsilon} G_{\mu\nu\eta\epsilon} \right) \\ & \left. - \sum_{i < j < k < l < m < n} J_{6\alpha} \left(\frac{1}{16} + \frac{1}{4} \sum_{\mu < \nu} \vec{S}_\mu \cdot \vec{S}_\nu + \sum_{\mu < \nu < \eta < \epsilon} G_{\mu\nu\eta\epsilon} + 4S_{ijklmn} \right) \right]. \end{aligned} \quad (1.25)$$

Another way to express the above Hamiltonian is to break exchange cycles into two-spin terms, four-spin terms..., and then sum each type of the two-spin terms. For example, in Fig. 1.3 (which is redrawn from Ref. [21] with the next-nearest-neighbor process J_{nnn} added), the three-particle exchange, J_t , can be broken into two nearest-neighbor terms, and one next-nearest-neighbor term. The four-particle cycle K_p can be divided into four nearest-neighbor terms or into one next-nearest-neighbor term and one third-nearest-neighbor term. Note that J_t and K_p are the notations used by Roger *et al.* [1] for particular exchange cycles in $J_{3\alpha}$ and $J_{4\alpha}$, respectively. See Refs. [20, 22] for the contributions of other exchange cycles. For some calculation, it is more convenient to express the Hamiltonian this way [20, 23]. Using the decoupling scheme with the constant terms removed for clarity, the Hamiltonian becomes

$$\begin{aligned}
 H_{exchange} = & -2J_1 \sum_{i < j}^{(1)} \vec{S}_i \cdot \vec{S}_j - 2J_2 \sum_{i < j}^{(2)} \vec{S}_i \cdot \vec{S}_j \cdots - 2J_k \sum_{i < j}^{(k)} \vec{S}_i \cdot \vec{S}_j - \cdots \\
 & + O(S^4) + O(S^6),
 \end{aligned} \tag{1.26}$$

where the four-spin term $O(S^4)$ has the form $(\vec{S}_i \cdot \vec{S}_j)(\vec{S}_k \cdot \vec{S}_l)$ and $O(S^6) = (\vec{S}_i \cdot \vec{S}_j)(\vec{S}_k \cdot \vec{S}_l)(\vec{S}_m \cdot \vec{S}_n)$. The summation $\sum^{(k)}$ is taken over the k^{th} nearest neighbor and J_k has the form $\sum_{n\alpha} c_{n\alpha} J_{n\alpha}$ with $c_{n\alpha}$ a constant. For example, it can be shown that for the exchange cycles given in Fig. 1.3

$$\begin{aligned}
 J_1 &= J_{nn} - 6J_t + 3K_p \\
 J_2 &= J_{nnn} - 4J_t + K_p \\
 J_3 &= \frac{K_p}{2}.
 \end{aligned} \tag{1.27}$$

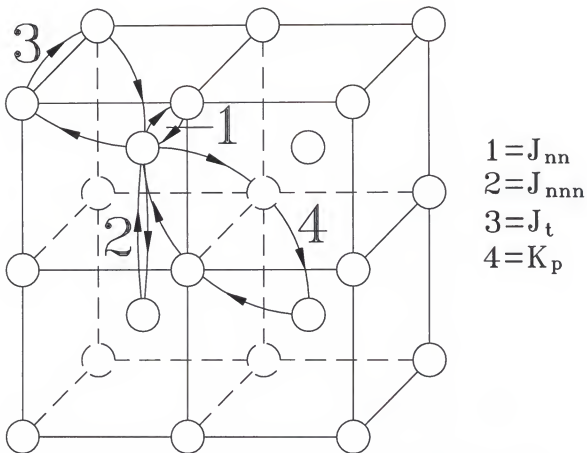


Figure 1.3. Schematic drawing of important exchange processes in bcc solid ^3He .

In 1983, Roger, Delrieu, and Hetherington [1] published a review paper in which they considered three and four particle exchange effects in the magnetism of bcc solid ^3He . Their goal was to find a Hamiltonian which gave results consistent with experimental data and at the same time contained the smallest number of exchange processes. They found that three- (J_t) and planar four-particle (K_p) exchanges would give reasonably good agreement with existing experimental data. The exchange Hamiltonian, Eq. 1.25, is reduced to only two terms

$$H_{\text{exchange}} = \sum_{i < j < k} J_t \left(\frac{1}{2} + 2(\vec{S}_i \cdot \vec{S}_j + \vec{S}_j \cdot \vec{S}_k + \vec{S}_k \cdot \vec{S}_i) \right) \quad (1.28)$$

$$- \sum_{i < j < k < l} K_p \left(\frac{1}{4} + \sum_{\mu < \nu} \vec{S}_\mu \cdot \vec{S}_\nu + 4G_{ijkl} \right).$$

As in the HNN model, this Hamiltonian was used to calculate the free energy (see Eq. 1.10). Various thermodynamic properties at high temperature were determined from the expansion of the free energy. For the bcc solid, Roger *et al.* [1] found

$$\theta_w = \frac{18}{k_B} (K_p - 2J_t), \quad (1.29)$$

$$e_2 = (24)^2 (J_t^2 - \frac{7}{8} K_p J_t + \frac{17}{64} K_p^2). \quad (1.30)$$

The expression for θ_w clearly shows the competition of three- (ferromagnetic) and four-particle (antiferromagnetic) exchanges. The values of J_t and K_p are obtained by fitting to the experimental results of the high-temperature coefficient e_2 and the mean spin-wave velocity v_{spin} which are found to be 7 (mK)² [24] and 8 cm/sec [25] respectively. The expression for v_{spin} used in the fitting is [1]

$$\begin{aligned} v_{spin} &= 2 \left(\frac{a}{\hbar} \right) (2J_1 + J_2 + 4J_3 + 3K_p)^{1/2} (-J_2)^{1/3} \left(J_2 + 4J_3 - \frac{J_1^2}{3K_p} \right)^{1/6} \\ &= 4 \frac{a}{\hbar} (3K_p - 4J_t)^{1/2} (4J_t - K_p)^{1/3} (8J_t - \frac{12J_t^2}{K_p})^{1/6}, \end{aligned} \quad (1.31)$$

where a is the lattice constant and J_i are given in Eq. 1.27 with $J_{nn} = J_{nnn} = 0$. For bcc solid at the melting density, J_t has a value of -0.13 mK and K_p of -0.385 mK yielding $\theta_w = -2.25$ mK and $T_N = 1.2$ mK (compared to the experimental value of 0.934 mK measured by Ni *et al.* [9]).

Using the two exchange model, Roger *et al.* were able to predict the U2D2 structure in the ordered low-field phase, a canted structure for the high-field phase, and a first-order transition line between the paramagnetic phase and the high-field phase in the low-field region. Although this is a much better model than the HNN model it also has weaknesses. For example, it predicts the critical field between the LFP and the HFP to be 1.6 Tesla at zero temperature. In reality $B_c(0)$ was measured to be 0.452 Tesla by Xia *et al.* [13]. To lower the critical field, Roger *et al.* introduced a third exchange process into the Hamiltonian. This process is the exchange of two nearest neighbors J_{nn} used in the HNN model. Unfortunately, this process creates an extra ordered phase in the phase diagram, which has not been observed experimentally.

In 1987, Ceperley and Jacucci [26] calculated the exchange frequencies of various processes using the path-integral Monte Carlo method. They found that even six-particle exchange processes are significant in bcc ^3He .

In the same year, Roger *et al.* [23] published the results of their high-temperature expansion with a multiple-exchange Hamiltonian. They included in the calculation two-, three-, and four-particle exchange processes for both bcc and hcp solids in an arbitrary field. The expansion was carried to fourth order in $\beta = 1/k_B T$, one order higher than the 1983 calculation [1]. Their exchange Hamiltonian has the form

$$H_{\text{Exchange}} = \tilde{J}_1 \sum_{i < j}^{(1)} P_{ij} + \tilde{J}_2 \sum_{i < j}^{(2)} P_{ij} + K \sum_{i < j < k < l} (P_{ijkl} + P_{ijkl}^{-1}), \quad (1.32)$$

where $\tilde{J}_1$, $\tilde{J}_2$ are the combination of two- and three-particle exchange as they appear in Eq. 1.27 and K is the exchange frequency involving four particles, with $\sum^{(1)}$ calculated over the nearest-neighbor pairs and $\sum^{(2)}$ over the next-nearest-neighbor pairs. For a bcc

solid, the expressions for the interactions are:

$$\begin{aligned}
 \tilde{J}_1 &= J_{nn} - 6J_t \\
 \tilde{J}_2 &= J_{nnn} - 4J_t \\
 K &= K_p.
 \end{aligned}
 \tag{1.33}$$

Note that $\tilde{J}_i$ does not include K_p ; therefore, it is not equal to J_i which is defined in Eq. 1.27.

The exchange processes are shown in Fig. 1.3.

For an hcp solid, Roger *et al.* [23] took into account five dominant exchange cycles: J_{nn} , J'_{nn} , J_t , J'_t , and K'_{sq} . These five dominant processes together with three less important exchange processes (J_t^* , K_P , and K'_P) are drawn in Fig. 1.4, which is taken from Okamoto's Ph.D. thesis [27] and modified slightly for clarity. The cross-hatched atoms are shown smaller in size to give a three-dimensional perspective. As in the bcc solid, the same expression for the exchange Hamiltonian, Eq. 1.32, is used but with

$$\begin{aligned}
 \tilde{J}_1 &= J_{nn} - J_t - 2J'_t, \\
 \tilde{J}_2 &= J'_{nn} - 4J'_t, \\
 K &= K'_{sq}.
 \end{aligned}
 \tag{1.34}$$

Due to the difference in the crystal structures, $\tilde{J}_i$ for a bcc and hcp solid are not the same. Note also that $\tilde{J}_2$ in the hcp case is for the nearest-neighbor pairs out of the basal plane instead of the next-nearest-neighbor pairs.

Once again, the free energy is calculated from the Hamiltonian and expanded in powers of β , similar to Eq. 1.11. Various thermodynamic quantities can then be obtained. For

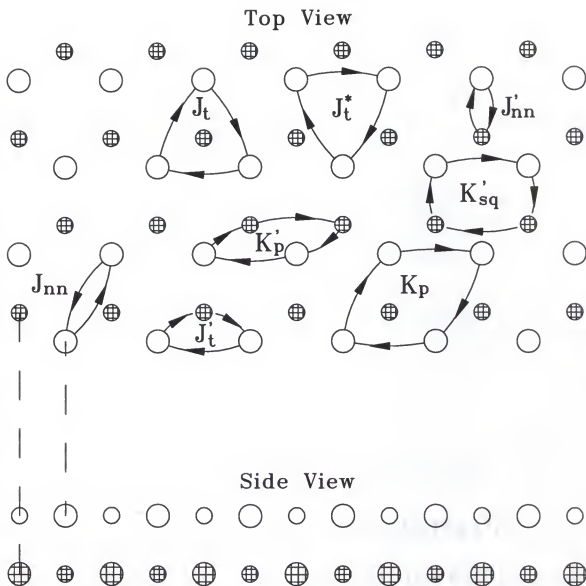


Figure 1.4. Schematic drawing of important particles exchange processes in hcp solid ^3He .

example, by taking the volume derivative of the free energy, up to the first-order term of $1/k_B T$ the pressure of the bcc solid is

$$P = R \left(\frac{\partial \theta_w}{\partial v} \frac{p^2}{2} + \frac{\partial K_p}{k_B \partial v} \frac{3p^4}{2} \right) + \dots, \quad (1.35)$$

and that for the hcp solid is

$$P = R \left(\frac{\partial \theta_w}{\partial v} \frac{p^2}{2} + \frac{\partial K'_{sq}}{k_B \partial v} \frac{3p^4}{4} \right) + \dots. \quad (1.36)$$

In the above equations, R is the gas constant, p the polarization⁴ which is given by $p = \tanh(\hbar \gamma B_0 / 2k_B T)$, and θ_w the Weiss temperature

$$k_B \theta_w = \begin{cases} 4\tilde{J}_1 + 3\tilde{J}_2 + 18K_p & \text{bcc solid} \\ 3\tilde{J}_1 + 3\tilde{J}_2 + 9K'_{sq} & \text{hcp solid.} \end{cases} \quad (1.37)$$

Since formulae 1.35 and 1.36 are valid for high magnetic field at high temperature, unlike Eq. 1.12, which requires $\hbar \gamma B_0 / k_B T \ll 1$, the volume derivatives of θ_w , K_p , and K'_{sq} can be calculated from the high-T pressure data of a ^3He sample (see Sections 4.1.1 and 4.2.1 for more discussion). Also, in Section 4.2.3 we have presented detailed description of how this model is used to simulate a sample demagnetization.

⁴For $p = 0$ half of the total number of spins are pointing in the same direction with the applied field and the other half are pointing in the opposite direction. For $p = 1$ all of the spins are pointing in the direction of the applied field.

1.3 Previous Experiments on Solid ^3He

1.3.1 bcc Solid

Because of the simple nature of ^3He , it was thought that the ordinary HNN model would explain all the magnetic properties of solid ^3He . Early high-temperature experimental data seemed to support this model quite well. However, in 1971, Kirk and Adams [28] made pressure measurements of various samples at different fields and found that there was a large discrepancy between the theoretical and experimental values of the Weiss temperature θ_w . This inconsistency yielded the first sign of problems with the simple HNN model. Below are brief descriptions of important experiments in the study of magnetism in solid ^3He .

In 1974, Halperin *et al.* [18] discovered the magnetic ordering transition in the bcc solid at melting density. At the transition temperature of about 1 mK, they found an abrupt decrease in entropy indicating that the transition is first order. In the recent measurement by Ni *et al.* [29], the entropies just above and below the transition temperature are found to be $0.63R\ln 2$ and $0.19R\ln 2$, respectively. The discovery by Halperin *et al.* contradicts the prediction by the HNN model of a second-order transition at 2 mK. The observation sparked much interest and many experiments soon followed. Realizing the insufficiency of the simple HNN model, theorists have sought better models which can explain the rich magnetic behavior of solid ^3He . One of these models is the two-exchange (J_t and K_p) model mentioned in the last section.

In 1977, Kummer *et al.* [30] carried out an experiment which was similar to the one performed by Halperin *et al.* but in a finite field up to 1.2 T. They found that the ordering transition decreased with increasing field up to a field of 0.4 T. With higher field the behavior was reversed, an increase in field raised the ordering temperature, causing a kink

in the phase diagram. They correctly identified the kink as an indication of a new ordering phase which is known today as the HFP (see Fig. 1.2).

The transitions to the LFP from HFP and PP are accepted to be first order. However, there were disputes about the order of the PP-HFP transition. An NMR experiment in 1981 by Osheroff [31] in fields up to 0.6 T indicated that the transition was first order. Similar conclusions were also drawn from two other works [32, 33]. In other experiments [34, 35], the results indicated second-order behavior. This disagreement was resolved in 1993 by Xia *et al.* [13]. In this work, they found that the transition was first-order for $B < 0.65$ Tesla, indicated by the discontinuity of the entropy across the transition. In higher fields, the entropy changed continuously through the phase transition, so here the transition is second order.

1.3.2 hcp Solid

While many significant works have been done on the bcc solid at low temperatures, there has not been much work on the hcp solid because the magnetic ordering of the hcp solid occurs at very low temperatures. An hcp solid near the bcc-hcp boundary is expected to order at about $20\ \mu\text{K}$ or less. The first attempt to cool an hcp sample into its ordered state was made in 1985 by Takano *et al.* [2]. Using PrNi_5 and copper nuclear demagnetization stages they cooled their samples to $43\ \mu\text{K}$, but no ordering signal was observed. However, by measuring the AC susceptibility of samples with molar volumes ranging from 18.1 to $19.4\ \text{cm}^3/\text{mole}$ and fitting the data to a Curie-Weiss form, $\chi = C/(T - \theta_w)$, they found a positive Weiss temperature θ_w for all samples. The result is an indication of a ferromagnetic tendency in the hcp solid. The fitting form is similar to the Eq. 1.14 but with the third term in the denominator set to zero in the high-temperature approximation.

A similar experiment was carried out by Yano *et al.* [3] in 1990. In this work, the samples were cooled to 38 μK by double nuclear demagnetization (PrNi_5 and Cu in series). Again, all Weiss temperatures were found to have a positive value, but there was no transition found in any of the samples. However, the molar volume dependence of θ_w was experimentally determined to be v^{18} , which was in good agreement with high temperature NMR data [8].

In 1989, Miura *et al.* [4] made the first pressure measurements on an hcp sample down to 0.93 mK in magnetic fields up to 0.2 T. Fitting the results to the high temperature expansion (Eq. 1.12) and using only $\partial\epsilon_2/\partial v$ and $\partial\theta_w/\partial v$ terms, they found $\partial\theta_w/\partial v$ to have a positive value, another indication of a ferromagnetic tendency in the hcp solid.

Even with the best cryostat available, current technology does not allow high-density solid ^3He to be indirectly cooled into its ordered phase by a nuclear demagnetization stage. In addition to the low ordering temperature, a long thermal time constant, which is proportional to $1/T$ in zero field [35], makes it essentially impossible to reach the ordering transition. Fortunately, direct demagnetization of solid ^3He nuclei (an idea that has existed for many years [5]) can bypass these problems. Recently, this method was used at the University of Tokyo by Okamoto *et al.* [6] in an attempt to cool ^3He solids, both bcc and hcp, into their ordered phases. This group successfully cooled a bcc sample with a volume of 19.99 cm^3/mole into its HFP and LFP phases. However, for their two hcp samples, with $v = 19.66$ and 19.07 cm^3/mole , respectively, they found no clear ordering transition.

One of the main problems with the Tokyo experiment was heating of the samples near the end of the demagnetization, attributed to an indium O-ring which became superconducting near zero field. To eliminate the problem, we have designed a better experimental cell in which there is no superconducting O-ring. The description of the cell is presented in Chapter 2 along with details of the experimental setup and techniques.

CHAPTER 2

EXPERIMENTAL APPARATUS

The objective of this experiment was to cool hcp ^3He to its magnetically ordered state and to determine the ordering by measuring the thermodynamic pressure. In order to cool an hcp sample into its magnetically ordered state, the sample itself was used as a second demagnetization stage. Our hcp data indicate that a sample temperature of $20\ \mu\text{K}$ or less can be achieved using this direct cooling method. The sample in a field of 2.5 T, along with a copper demagnetization stage in a field of 7 T, was precooled to 11 mK by a dilution refrigerator. Then, the copper stage was demagnetized to lower the temperature. Upon reaching the desired temperature, the sample was demagnetized to zero field. During the sample precooling process, the temperature of the copper stage was measured by a ^3He melting pressure thermometer while the conditions of the dilution refrigerator were monitored by various resistance thermometers.

The pressure change in the solid was detected by measuring the change in the capacitance of a strain gauge. Since the pressure change was small (on order of 10 Pa) and the measurements were made at microkelvin temperatures with a changing magnetic field, there were many restrictions in the designs of the pressure gauge and the cell. Nevertheless, with a new design of the cell, we have avoided significant heating encountered by the Tokyo group [6] near the end of the sample demagnetization resulting in a better understanding of the pressure data and a clear observation of the magnetic ordering in hcp solid.

In this chapter, the experimental apparatus used in this work is covered in detail. The first part describes the experimental cell which was used to hold the sample and to detect the pressure change. Two different experimental cells were made for this work, the second cell being an improved version of the first. The main goals for the second design were to minimize the magnetic-field-dependence and maximize the thermal conductance of the cell. The first section deals exclusively with the second cell in which magnetic ordering of the bcc and hcp solids was observed. The second section describes the electronic set-up of the bridge used to detect capacitance changes in the strain gauge of the cell. The third section details the construction of a 2.5 T home-made magnet which was used to demagnetize the ^3He sample. The fourth section describes the cryostat and the fifth section details the thermometric methods employed.

2.1 Experimental Cell

A schematic drawing of the experimental cell is shown in Fig. 2.1. The pressure change in the sample was detected by a capacitive pressure transducer of simple design. One wall of the sample cell is a thin, flexible diaphragm which moves up and down in response to increases and decreases in the pressure of the sample. A coin-silver disk is attached to the diaphragm and serves as a movable capacitor plate while another disk, held close to the movable plate, is used as a fixed plate. As the pressure changes, the gap between the two plates varies, leading to changes in capacitance. The magnitude of the change can be detected by a capacitance bridge, the design and operation of which is described in the next section.

Taking pressure measurements during the precooling and nuclear demagnetization of high density solid ^3He requires two main considerations in the design of the cell: the

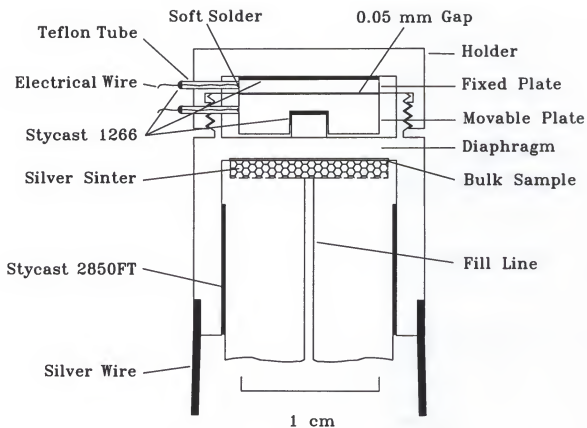


Figure 2.1. Schematic drawing of the experimental cell.

maximization of the thermal conductance between the sample, the cell and the copper nuclear stage and the reduction of eddy-current heating which exists during the sample demagnetization. While the first condition calls for a large area in the design of the cell, the second requires a small cross-sectional area. Both of the constraints are met in the design of this cell, as described in the following text.

The upper part of the cell, made of coin silver (90% Ag and 10% Cu), is a Straty-Adams strain gauge which has a fixed-edge diaphragm.¹ The diaphragm of the gauge has a diameter of 1.22 cm (0.48") and thickness 1.6 mm (0.063") chosen so that the strain gauge can withstand high pressure and at the same time yield high sensitivity. The formula used for this determination can be found in a review article by Adams[36] and is given by

$$t = \sqrt{\frac{3a^2 P_m}{4S_m}}, \quad (2.1)$$

where t is the thickness, a the radius of the diaphragm, P_m the maximum operating pressure and S_m the maximum stress. For this cell, S_m was taken to be 150 MPa (21000 psi) and P_m was 15 MPa (2100 psi).

Learning from the first gauge that the mylar spacer and stainless steel screws had caused a large heat load and magnetic-field-dependence, the second gauge was designed without them. The screws were no longer needed because the holder for the fixed plate had threads on its inner wall, similar to the cap of a jar, while the other holder had similar threads on its outer wall. These were machined so that when they were tightly screwed together there was a 0.05 mm (0.002") gap between the two plates which eliminated the need for a spacer. The gauge was thermally linked to the rest of the cell by two annealed silver wires 1.3 mm

¹The diaphragm is machined into the sample cell so that its edge is held fixed by the wall of the cell.

(0.05") in diameter which were welded to the holder of the movable plate at one end and to small silver blocks at the other. The blocks, not shown in the figure, were tightly fixed to the base of the cell by stainless steel screws. To reduce eddy current heating during the sample demagnetization, the holder of the fixed plate and the two plates had slots cut into them. The use of the annealed silver wires together with the presence of slots at various positions on the cell enable the two constraints of maximizing thermal conductance and minimizing eddy-current heating to be satisfied.

Electrical wires were soldered to the plates using EutecRod #157 soft solder.² To protect the solder joints, the wires were placed inside short pieces of teflon tube and a small amount of Stycast 1266 epoxy³ was applied to hold the tube in place. The wires were single filament superconducting wires, the same type used for winding the 1 T magnet.⁴ The two capacitor plates were glued to their holders with Stycast 1266 epoxy using thin porous Kimwipe paper wetted with Stycast to provide electrical insulation. After the Stycast had dried, the plates were re-machined together with their holders to ensure that they were parallel to one another.

The lower part of the cell, made of silver of 99.999% purity, was a cylinder with diameter of 1.3 cm (0.5") and length of 7.4 cm (2.9"). A shallow flat-bottomed hole was bored in the top end with seven shallow holes added at the bottom of the counter-bore to provide a stronger bond between the silver powder and the cylinder. Two slots of dimensions 0.05 cm x 4.8 cm (0.02" x 1.9") were cut into the body of the cylinder and at right angles to each other in order to reduce eddy current heating. The bottom end of the cylinder was welded to a silver plate of thickness of 0.31 cm (0.125") with 11 clearance holes for screws

²Eutetic Corporation, 4040 172nd St., Flushing, NY 11358, phone: (800) 323-4845.

³Deanco, 7130 University Blvd., Winter Park, FL 32792, phone: (407) 671-5100.

⁴To ensure that they would remain superconducting in high fields and thus generate less heat during the pressure measurements.

which were used to clamp the cell to the copper nuclear demagnetization stage. Slots were cut between these holes to maximize the contact area between the cell and copper stage by allowing the silver plate to flex. Without the slots, the rigid base of the cell could have as few as three contact points to the copper stage.

The lower part of the cell was annealed for 12 hours at 750°C (1380°F) with a pressure of 10^{-4} Torr of O₂ gas. To reduce the contamination from oil mist and other foreign materials inside the oven, greened stainless steel sheets were used to shield the silver. After annealing, the silver was left inside the oven to cool to room temperature over night. A residual resistance ratio (RRR) of about 400 was achieved using this procedure. A copper-nickel (Cu-Ni) fill line with a diameter of 5 mm (0.02") was soft soldered to the body of the cell 1.59 cm (0.63") below the sinter area using EutecRod #157 solder.

Silver sinter,⁵ of 700 Å particles pre-heated at 295°C, was then packed in the cell to provide good thermal contact between the bulk ³He and the rest of the cell. During the packing process, a packing pressure of 6.9 MPa (1000 psi) was applied to a small amount of sinter which was added at each step to ensure the same density throughout the cell. The final packing ratio of 50% yielded a surface area of approximately 1 m², for the 0.5 g of sinter powder.

The heating problem encountered by Okamoto *et al.* [6] near the end of their sample demagnetization was caused by flux jumps in a superconducting indium O-ring. The best way to solve this problem is to completely eliminate the indium ring from the structure of the cell. This was achieved in the design of our cell by replacing the indium with an epoxy seal.

⁵ULVAC Technologies Inc., 6 Riverside Dr., Andover, MA 01810, phone: (508) 686-7550, fax: (508) 689-6300.

The strain gauge and the body of the cell were held together and vacuum sealed by Stycast 2850FT epoxy prepared with low viscosity catalyst 24 LV. This catalyst was chosen because it has better adhesion and higher resistance to thermal shock than catalyst 9 or 11. The epoxy was left to cure for about 12 hours at room temperature, as recommended by the manufacturer. The cell was leak-checked at liquid Nitrogen (LN_2) temperature with a pressure up to 15MPa (2200 psi), safely above the pressure 14MPa (2000 psi) at which the low density hcp solid forms.

2.2 Pressure-Measurement Method

The capacitance change in the strain gauge was detected using a bridge configuration in which the unknown capacitance of the strain gauge was compared to a reference capacitor. The reference capacitor, made from beryllium-copper (BeCu), was designed similar to the experimental strain gauge. The plates of the capacitor were glued to their holders using Stycast epoxy, which were separated by a mylar spacer with a thickness of 0.05 mm (0.002") and bolted together to achieve mechanical stability.

For high-precision measurements, the reference capacitor must be stable, $\delta C/C < 10^{-7}$, a condition which is almost impossible to meet at room temperature.⁶ Fortunately, stability can be achieved by placing the capacitor on the mixing chamber of the dilution refrigerator. In the millikelvin regime the thermal expansion coefficient of BeCu is small so that any change in the temperature of the reference capacitor does not create a detectable change in its capacitance.

Three coaxial lines were used in this electronic arrangement. The strain gauge and reference capacitor each had their own isolated line, and the third line was common to

⁶Because the large thermal expansion of materials at high temperatures means that the temperature of the capacitor would have to be kept extremely stable.

both. Below the 1 K pot the coaxial cables were constructed by inserting niobium-titanium (NbTi) superconducting wires inside CuNi capillaries. Before insertion, the wires were coated with vacuum grease to keep them rigid inside the tubes. The cables were thermally anchored to various parts of the refrigerator, such as the 1 K pot and mixing chamber. To improve the heat sinking of the coaxes, the thermal contact area of the tubes was increased by wrapping the tubes around Cu formers and soft soldering them together. At the top of the cryostat the lines were terminated inside a vacuum-tight brass header.

A schematic of the bridge is shown in Fig. 2.2. The Eaton 1011A ratio standard⁷ and capacitors were isolated from ground by a 1:1 Eaton ST-200A isolation transformer. Referring to the figure, C_p denotes the unknown capacitance of the pressure gauge of the experimental cell, and C_{Ref} the value of the reference capacitor. A variable resistor was used to balance the resistive part of the signal. The resistor had a maximum value of 100 Ω and was placed in an aluminum box to shield rf noise.

A personal computer (316S CompuAdd⁸) was used to control the output of the HP 3325A function generator⁹ and to read and store data. A low noise pre-amplifier (Ithaco 1201¹⁰) was added to improve the signal-to-noise ratio. The output signal, V_0 , was amplified by 10^4 before being fed to the PAR 124A lock-in amplifier.¹¹ In addition, the high pass and low pass rolloffs of the pre-amplifier were set at 100 Hz and 1000 Hz, respectively, to narrow the bandwidth. The pre-amplifier was connected to the coaxial header on top of the cryostat via a short coaxial cable and enclosed in a polystyrene box to protect it from room

⁷Eaton Corporation, Electronic Instrumentation Division, 5340 Alla Road, Los Angeles, CA 90066, phone: (213) 822-3061.

⁸CompuAdd, 12303-G Technology Blvd, P.O. Box 200777, Austin, TX 78727-0777, phone: (800) 999-9901.

⁹Hewlett-Packard, P. O. Box 105005, Atlanta, GA 30348, phone: (800) 452-4844, fax: (404) 980-7256.

¹⁰Ithaco, Inc., 735 West Clinton Street, P. O. Box 6437, Ithaca, NY 14851-6437, phone: (607) 272-7640, fax: (607) 272-4550.

¹¹EG&G Princeton Applied Research, P. O. Box 2565, Princeton, NJ 08643-2565, phone: (512) 272-9621 or (512) 272-9649.

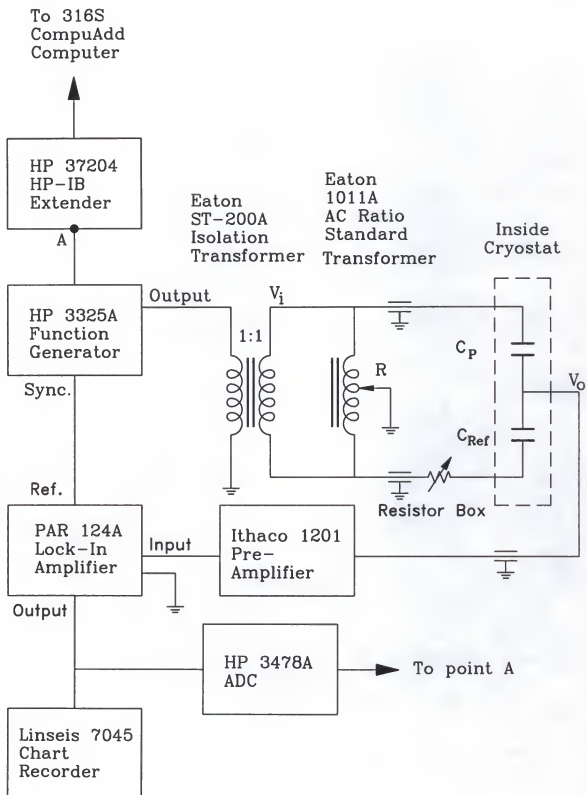


Figure 2.2. Electronic circuit of the capacitance bridge.

temperature fluctuations. The in-phase signal of the lock-in amplifier was recorded by a Linseis chart recorder¹² and a Hewlett-Packard HP 3478A multimeter digitized the output signal for storing in the computer.

Except for the computer, everything shown in Fig. 2.2 was located inside the shielded room. Communication between the computer and the shielded-room electronics was made through two HP 37204 HP-IB extenders which were placed at the two ends of the communication line and linked together by a fiber-optic cable in order to eliminate rf noise. The equipment and the computer were connected to the extenders with GPIB cables.

For an ideal circuit, the input from the function generator and the output voltage of the bridge are related by:

$$\frac{V_0}{V_i} = \frac{(R-1)C_p + RC_{Ref}}{C_p + C_{Ref}}, \quad (2.2)$$

where R is the value of the ratio standard. When the bridge is balanced, $V_0 = 0$, the equation reduces to become

$$\frac{C_p}{C_{Ref}} = \frac{R}{1-R}. \quad (2.3)$$

If C_{Ref} is known, the value of C_p can be determined using the balance point R . However, the value of C_p is not needed in calculating the pressure change of the sample because, through a calibration, the pressure can be expressed as a function of R , as discussed in a later part of this section.

In a more realistic model, it can be shown that a large capacitance in the output cable can significantly reduce the bridge sensitivity [36]. This problem can be minimized by using a short length of cable. Further analysis of the model reveals that the change in

¹²Linseis, Inc., P. O. Box 666, Princeton Jct., NJ 08550-0660, phone: (800) 732-6733.

out-of-phase component can be minimized by having C_p equal to C_{Ref} and similar cable capacitance values.

To minimize heating of the capacitor plates of the strain gauge, the output of the function generator (HP3325A) was turned on only when a measurement was made. The HP3325A was used instead of the internal oscillator of the PAR124A lock-in amplifier because its output could be controlled by a computer. The on/off timing of the HP3325A was remotely controlled by a LabWindow program which was run on the 316S CompuAdd. The program was written by Dr. D. A. Sergatskov and was designed to simultaneously control three HP devices: the HP 3325A Function Generator, the HP 3478A Multimeter, and the HP 3457A Multimeter (not shown in Fig. 2.2).

When running, the program first sent out a command to turn on the output of the HP 3325A function generator. The output signal was a $2.12\text{-}V_{pp}$ sine wave at 317 Hz whose amplitude and frequency could be set in the program menu. The frequency was chosen to avoid coupling to the 60 Hz frequency of the power line. With an excitation of $2.12\text{ }V_{pp}$, the sensitivity of the cell was approximately 5 Pa (0.0007 psi) at 11 MPa (1600 psi). Higher excitation levels increased the signal-to-noise ratio but also generated more heating in the plates, which was not desirable.

Next the program read the outputs of the HP 3478A and HP 3457A multimeters. The HP 3478A multimeter was used exclusively for the experimental cell. Depending on the stage of the experiment, the HP 3457A was used for various devices such as the melting pressure, RuO_2 resistance, and Pt NMR thermometers, or measurement of current in the sample magnet. A typical measurement had a duration of 90 seconds, during which time the program read and stored data points in the computer memory at a rate of 2 points/sec.

Finally, the program switched off the HP 3325A by setting the output voltage level to zero. The program averaged the last 20 read points (10 seconds) and stored the value in a computer file. Only these last points were reliable because of the 30 second time constant, τ , set on the PAR 124A. The lock-in amplifier's output yields correct readings only after the signal has been turned on for an amount of time greater than about three τ .

During the precooling of a sample (which could last as long as six weeks), one measurement with a duration of 90 seconds was made every hour to monitor the slow change in pressure. In a 34 hour sample demagnetization, the waiting time between two measurements was reduced from 1 hour to 10 minutes, and then to 5 minutes for fields below 0.1 T in order to obtain better resolution of the pressure change of the sample with magnetic field. Once the onset of ordering was observed, data were taken continuously in order to provide more data below the transition point since the magnetic transition occurs at very low field. With a demagnetization rate of 73 mT/hr, the sample demagnetization below the transition was completed in a few minutes.

2.3 Superconducting Magnet

In this work, two superconducting magnets were made with maximum fields of 1 T and 2.5 T, respectively, in vacuum.¹³ The main difference between them is the type of superconducting wire used. The first magnet (1 T) has a single filament superconducting wire, and the second (2.5 T) a multi-filament wire, the construction of which is described in the following text.

¹³The 1 T magnet was initially used in precooling of the ^3He sample but was replaced by the 2.5 T magnet in order to raise the precooling temperature.

The magnet wire, made by Supercon, Inc.,¹⁴ has 54 superconducting filaments of 52% niobium-48% titanium embedded in a 0.257 mm (0.0101") dia. copper wire. The wire is electrically insulated by a layer of Formvar and has a total diameter of 0.287 mm (0.0113").

The magnet consists of two parts as shown in Fig. 2.3 and is based on the design of Isareleson and Gould [37]. The first part consists of a main coil, two notches, and two vertical compensation coils. The notches increase the uniformity of the field near the center of the magnet and the vertical compensation coils, wound in the opposite direction of the main coil, reduce the stray field along the z-axis. The suppression of the vertical field outside the magnet is particularly important because an aluminum superconducting heat switch with a critical field of only 11 mT is located directly above the magnet and any large stray field would reduce its effectiveness.

The main coil consisted of 21 layers, each having 264 turns. The notches each had 5 layers with 71 turns per layer and the vertical compensation coils had 14 layers with 35 turns per layer. Upon completion each layer was brushed with a dilute solution of GE varnish and acetone to prevent it from loosening. When the winding of the first part was completed, the outer layer was coated with Stycast 1266 epoxy. The whole part was then immediately placed into a dessicator and pumped to remove air so that the epoxy could penetrate into the inner layers.

The second part is an active shield which has 3 layers (wound in the opposite direction of the main coil) with 372 turns in the first layer, 321 turns in the second layer, and 270 turns in the third. Like the first part, it was coated with dilute GE varnish, and epoxy and an additional layer of dental floss was added to provide extra protection for the outermost

¹⁴Supercon, Inc., 830 Boston Turnpike, Shrewsbury, MA 01545, phone: (508) 842-0174, fax: (508) 842-0847.

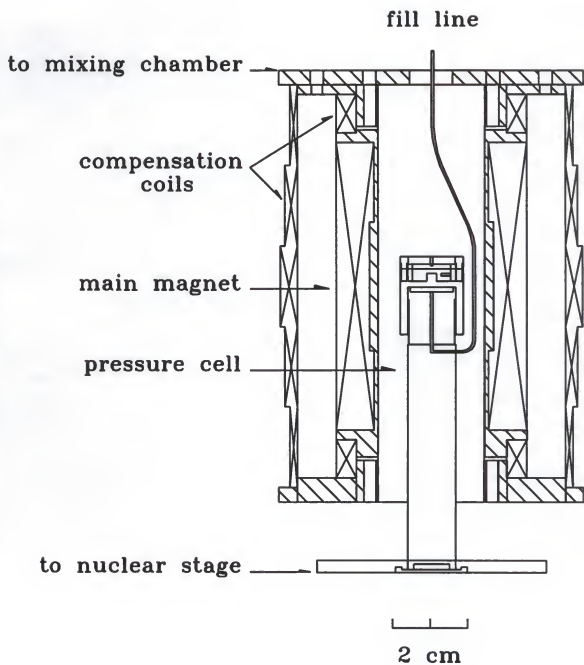


Figure 2.3. Experimental cell and magnet.

layer. The shield was made in an attempt to cancel out the stray field in the radial direction, but was not very successful. With a center field in excess of 0.73 T, the outside field was large enough to prevent the nearby Pt NMR thermometer from working. Its resonant signal was completely destroyed by the stray field, despite having a superconducting lead shield surrounding it. With a 0.73 T center field, the stray field at the thermometer was estimated to be 20 mT. However, this was large enough to interfere with the 14 mT operating field of the thermometer. With space limitations and a large field requirement for the precooling of the sample, the problem was difficult to solve and meant that the Pt NMR thermometer could not be used in this work. However, the lack of the Pt NMR thermometer did not create a significant set back to the experiment since the temperature of the bundle could be measured by a melting pressure thermometer down to 0.4 mK,¹⁵ described in Section 2.5.1. Lower temperatures could be estimated by using the magnetic field on the Cu bundle and comparing it to the values in other demagnetizations in which the Pt NMR thermometer had been used.

The bore of the magnet has a diameter of 2.84 cm (1.12"). Its large diameter is designed not only to work with the current pressure cell but also with future susceptibility experiments which will require a large space. Including the active shield, the total size of the magnet is a cylinder of diameter 8.1 cm (3.2") and length 11.4 cm (4.5"). The magnet was thermally anchored to the mixing chamber and supported by three Vespel-22 legs¹⁶ to prevent a heat leak to the copper stage. The legs have a diameter of 0.64 cm (0.25") and a length of 1.8 cm (0.7"). Figure 2.4 shows the schematic of the experimental arrangement.

¹⁵The lowest temperature needed in this experiment was 0.5 mK for 100% entropy removal in a sample.

¹⁶E. I. Du Pont De Nemours & Company (Inc.), Du Pont Polymers, Wilmington, DE 19898, phone: (800) - 222-8377.

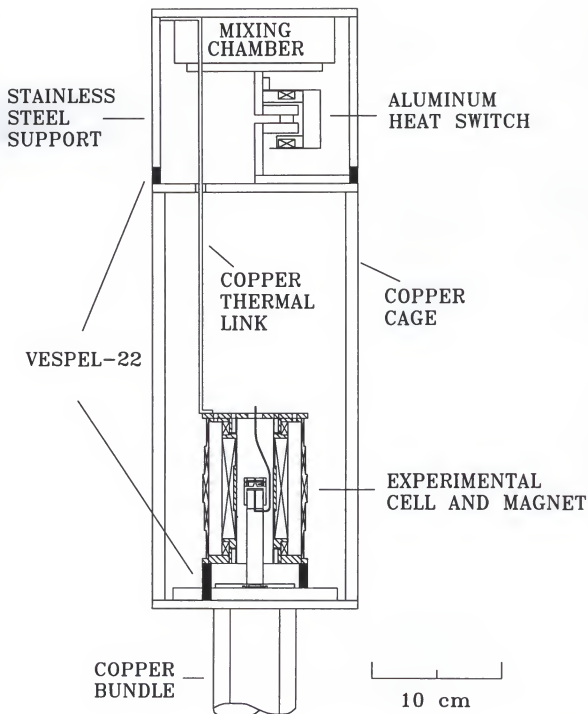


Figure 2.4. Experimental arrangement.

To help design the magnet a computer program written in FORTRAN by Chris Gould at the University of Southern California was used to calculate the field profiles. It allows the user to specify the dimensions of the coils and their locations with input parameters of the coil inner radii, outer radii, half length values, vertical offsets to the main coil, and the current density. Dennis Greywall, at AT&T Bell Lab, also wrote a similar program and distributed it as an executable file. Except for its inability to handle the vertical compensation coils, it can calculate the field profile for a magnet which consists of a main coil, notches, and an active shield. The Greywall program is easier to use and was employed in designing simple magnets such as that of the heat switch. Figure 2.5 shows how well the calculated curve, which has been generated by Gould's program, agrees with data along the z-axis of the magnet measured at 4 K using a Hall probe.

The operation of the superconducting magnet requires a persistent switch [38] whose function is to keep a current in the superconducting magnet without having it connected to a power supply at all times. The switch, located on top of the vacuum can, reduces heating and noise dumped into the cryostat. The construction of the switch is quite simple and consists of three main parts: a stainless steel tube, a segment of superconducting wire, and a heater wire. The stainless steel tube has a length of 5 cm (2") with a 0.64 cm (0.25") diameter and a wall thickness of 0.5 mm (0.02"). The superconducting wire is multi-filament 52% niobium-48% titanium, the same type of wire that is used for the magnet. Its length is roughly 1.2 m (4 ft) allowing both of its ends to reach the mixing chamber where they can be clamped together with the wires of the 2.5 T magnet. The Cu-Ni heater wire is 61 cm long and 0.08 mm (0.003") in diameter with a resistance of 2.86 Ω /cm (87.08 Ω /ft). The heater wire was tightly wound non-inductively around the superconducting wire as shown in Fig. 2.6.

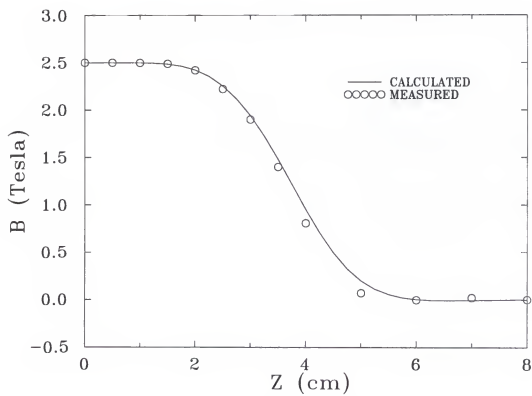


Figure 2.5. Field profile of the 2.5 Tesla magnet.

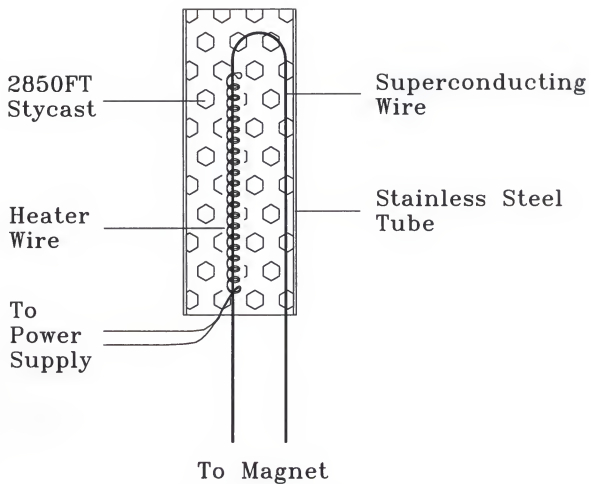


Figure 2.6. Persistent switch for superconducting magnet.

The superconducting wire was bent carefully, to prevent making a kink, at its mid-point. Both wires were inserted inside the stainless steel tube and Stycast 2850FT epoxy was used to fill the empty space. The epoxy keeps the wires from vibrating and provides thermal insulation. The persistent switch requires a current of approximately 20 mA to keep the superconducting wire in the normal state.

The switch was tied down on the top of the vacuum can with the superconducting wire run inside the can down to the mixing chamber. The wire was carefully thermal anchored at various parts of the refrigerator (such as the 1 K pot and still) using GE varnish, silver tape, and dental floss. At the mixing chamber, each end of the wire was connected to the magnet leads by clamping with the following method.

Before the wires were clamped, about 5 cm (2") of Formvar insulation was stripped from each end by Strip-X.¹⁷ The copper layer was then etched away with a dilute nitric acid solution until the superconducting filaments were completely exposed. The filaments from the magnet and the persistent switch were twisted together and inserted into a Cu tube approximately 6 cm (2.5") in length and 0.5 cm (0.2") in diameter which was then flattened using a press with a pressure of 660 MPa. With such a great pressure, an impression of the filaments could be seen on the outside of the flattened copper tubes. It was important to keep the pressure below 1 GPa to prevent stretching of the copper and causing the filaments to break. Using this type of clamp a test on two straight wires showed that the filaments remained superconducting with current up to 100 A in liquid ^4He . The clamping of the magnet leads worked extremely well: after six weeks of persisting at 2.5 T (34 A) during a long sample precooling, the decay of current in the magnet was not detectable.

¹⁷GC Electronics, 1801 Morgan Street, Rockford, IL 61102.

To energize the magnet, the heater of the persistent switch is turned on (step A of Fig. 2.7) causing the superconducting segment to become normal. The normal segment therefore has a large resistance compared with the superconducting magnet wire so that all current from the power supply flows directly to the magnet. The current in the magnet can then be increased, and when the maximum current is reached, the heater is turned off. The wire segment becomes superconducting and the current begins to flow through the switch (step B). Initially, the currents from the power supply and the magnet are equal, so the net current in the switch is zero. However, as the current supply is ramped down, the current that flows through the switch increases (step C). Finally the magnet current is persisted when there is no current remaining in the power supply (step D).

To remove the current from the magnet, the steps in Fig. 2.7 are carried out in reverse order, from D to A. The current inside the magnet must first be matched by that of the outside power supply. The current inside the switch gets smaller, step C, and eventually becomes zero when the currents have the same value, step B. The heater is then turned on, step A, and the current in the magnet can be removed by reducing the current in the power supply.

The magnet was energized by a programable 100 A Kepco ATE6-100A power supply¹⁸ whose output current can be controlled remotely by applying a dc voltage between 0 and 1 V to its control terminals. A ramp unit built by our electronics shop automatically controls the ramping process. It is a digital linear waveform generator which consists of two main parts. The first is a microcontroller board which allows the user to input the target current and ramp time via a computer terminal and an RS232 cable. The second part is a 16 bit digital-to-analog converter (DAC) board whose function is to convert the

¹⁸Kepco, Inc., 131-38 Sanford Avenue, Flushing, NY 11352, phone: (718) 461-7000.

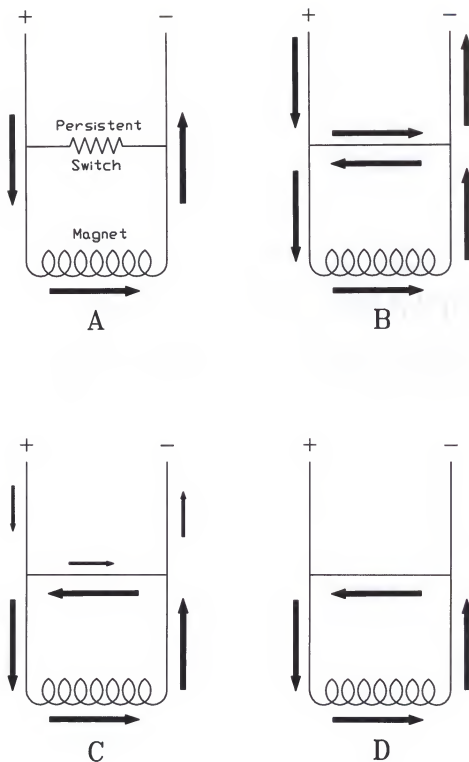


Figure 2.7. Steps in ramping up and persisting current in a superconducting magnet, see text for more detail.

digital signal from the microcontroller to an analog control voltage for the power supply. The full output range of the ramp unit is from 0 to 5 V and a voltage divider is used to match the control voltage to that of the power supply.

2.4 Cryostat

The cryostat is suspended from an aluminum hexagonal plate with side length 0.70 m (27.75") and thickness 7.6 cm (3") consisting of three layers. The top and bottom layers are made of solid aluminum and each has a thickness of 1.3 cm (0.5") while the middle layer, which is bonded with epoxy to the outer layers, has an aluminum honeycomb structure with thickness 5.1 cm (2.0"). With this type of construction the plate is light but strong enough to support the weight of the cryostat without bending. The plate is bolted on three hollow aluminum square beams which are welded together to form an equilateral triangle. Three Vibraplane 1206 active-air isolation mounts, made by Kinetic Systems, Inc.,¹⁹ support the triangle at its corners. The mounts are set on top of a 4-m-(13') tall concrete tripod which sits on three 2-ton concrete blocks. The blocks are isolated from building vibrations by soft rubber gaskets. This arrangement helps to minimize mechanical vibration which might couple to the cryostat. For more information, see Ref. [39].

The cryostat can be divided roughly into two main cooling stages. The first part is the dilution refrigerator and the second part is the copper demagnetization stage. An aluminum superconducting heat switch, situated between the two parts, controls the heat flow between them. Additional information on the cryostat can also be found in Ref. [40].

¹⁹Kinetic Systems, Inc., 20 Arboretum Road, P.O. Box K, Roslindale, MA 02131, phone: (617) 522-8700, fax: (617) 522-6323.

2.4.1 Dilution Refrigerator

The dilution unit is an Oxford Instruments model 1000.²⁰ The volume of the $^3\text{He}/^4\text{He}$ mixture used in this unit is approximately 250 NTP liters at room temperature with 16.4% of ^3He .²¹ The mixture is stored in three banks of storage tanks each of which has a capacity of 117 l.

The gas circulation system of the unit is composed of two Edwards 9B3 diffusion pumps,²² an Edwards EH250 mechanical booster (Roots), and an Alcatel 2063H rotary pump²³ connected in series in the cited order. The 9B3's are separated from the still line by an air-operated gate valve and a freon-cooled baffle which prevents mist of the pump oil from getting into the cryostat.

Figure 2.8 shows the gas handling system of the cryostat where dashed lines represent hidden gas lines. Valve #10 is a relief solenoid valve which automatically opens when the line pressure exceeds 90 KPa (650 Torr). Valve #32 is a one-way safety valve and #23 and #36 are micro-valves. When the dilution unit is running, the ^3He with about 10% ^4He is pumped from the still (F) by the 9B3's. The mixture circulates through the Roots and Alcatel pumps and returns to the gas panel from the back side of the Alcatel. Before going back to the cryostat, the mixture goes through one of the two LN_2 cold traps, a Hasting U5KM flow meter,²⁴ and valve #29.

²⁰Oxford Instruments, 130A Baker Avenue, Concord, MA 01742, phone: (508) 369-9933, fax: (508) 369-6616.

²¹This data is obtained from measurement made on 3/2/1990 during testing #3 of the cryostat.

²²Edwards High Vacuum International, One Edwards Park, 301 Ballardvale Street, Wilmington, MA 01887, phone: (800) 848-9800, fax: (508) 658-7969.

²³Alcatel, 40 Pond Park Road, South Shore Park, Hingham, MA 02043, phone: (617) 749-8710, fax: (617)-749-8660.

²⁴Teledyne Brown Engineering, Hasting Instruments, P. O. Box 1436, Hampton, VA 23661, phone: (800) 950-2468, fax: (804) 723-3925.

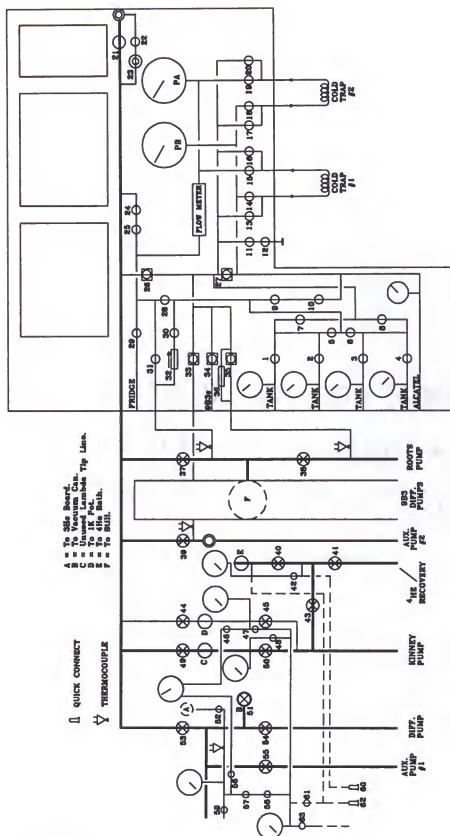


Figure 2.8. Gas handling system of the cryostat.

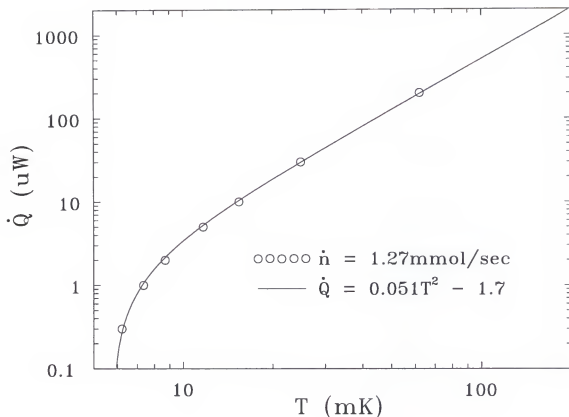


Figure 2.9. Cooling power of the Oxford 1000 dilution unit.

Figure 2.9 shows the cooling power of the unit from 110 mK down to 5 mK taken from measurements made in October of 1992 after the removal of some coaxial cables which had caused a large heat leak to the mixing chamber. The circulation rate is that of the whole mixture, measuring ^3He and ^4He combined. The cooling power of the mixing chamber can be expressed by [41]

$$dQ/dt = aT_{mc}^2 - b \quad (2.4)$$

where T_{mc} is the temperature of the mixing chamber and a and b the fitting parameters, except near the minimum temperature where the fit is not valid.

2.4.2 Aluminum Superconducting Heat Switch

The copper bundle can be thermally isolated from the dilution unit with an aluminum superconducting heat switch. Aluminum becomes superconducting at about 1.2 K in zero field and has a critical field $B_c(0)$ of about 11 mT. The thermal conductivity of aluminum is electronic and is thus linear with temperature in the normal state (κ_n), with a T^3 dependence when it becomes superconducting (κ_s). For $T < 1.2$ K, the switch can be closed (normal) by applying a field greater than B_c or opened (superconducting) by removing the applied field. In 1978, Mueller *et al.* [42] constructed the first aluminum heat switch. Their switch has a large switching ratio κ_n/κ_s of $1600/T^2$ at 58 mK compared to the typical values of $100/T^2$ to $300/T^2$ of a more popular tin (Sn) switch. One main problem encountered by Mueller *et al.* in making an aluminum heat switch was that its surface becomes oxidized very quickly when exposed to air. This problem was solved by electroplating the aluminum with gold, a procedure described in Ref. [42].

In precooling of the copper bundle the cooling rate depends on the thermal conductance between the mixing chamber and the bundle. For this cryostat we found that conductance between the contact of the heat switch and the thread annulus of the mixing chamber was poor ($105T$ mW/K²), a serious problem in the design of the mixing chamber by Oxford. This problem was overcome by constructing a heat switch which can be bolted to 24 available tapped holes at the bottom of the mixing chamber. This arrangement provided a measured conductance of $540T$ mW/K², an improvement over Oxford's design by a factor of 5.

The conductance of the heat switch itself was also improved from 210 to $390T$ mW/K² by increasing the thickness of the aluminum strips from 0.5 mm to 1 mm. With these two

improvements, the net conductance between the mixing chamber and the copper bundle was increased from 61 to 200 T mW/K² which reduces the cooling time by half [40].

2.4.3 Copper Demagnetization Stage

The stage is made from 350 moles of Cu consisting of 19 Cu plates,²⁵ each of which has a thickness of 3.2 mm (0.125"). The plates have a length of 91 cm (36") and a width varying from 11 mm (0.43") to 63 mm (2.5"). When put together, the plates have the form of a cylinder of 63 mm (2.5") in diameter, see Fig. 2.10. The plates are electron-beam welded to copper flanges at each end. The flanges are gold plated to reduce oxidation and provide good thermal contact for attaching experiments.

The performance of the dilution unit during a precooling of the copper nuclear demagnetization stage is shown in Fig. 2.11. The expression for the precooling time t required for the stage to reach a certain temperature T can be derived in the following way. First, the heat flow from the copper stage to the mixing chamber is given by

$$dQ/dt = \frac{k}{2}(T^2 - T_{mc}^2). \quad (2.5)$$

Using Eq. 2.4 to eliminate T_{mc}^2 , one obtains

$$dQ/dt = \frac{\frac{k}{2}}{\frac{k}{2} + a}(aT^2 - b). \quad (2.6)$$

The nuclear heat capacity of the copper stage has the form

$$C = \frac{c}{T^2} \quad (2.7)$$

²⁵With 173 of the 350 moles is in a 8 T magnet. The use of many plates in place of a single piece is to minimize the cross-section area of the copper stage and thus reduce the eddy-current heating during a demagnetization.

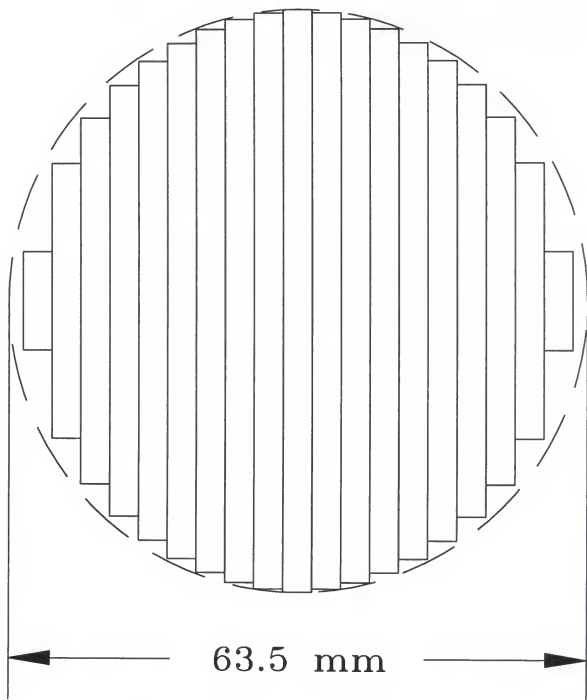


Figure 2.10. Cross-section view of the copper bundle.

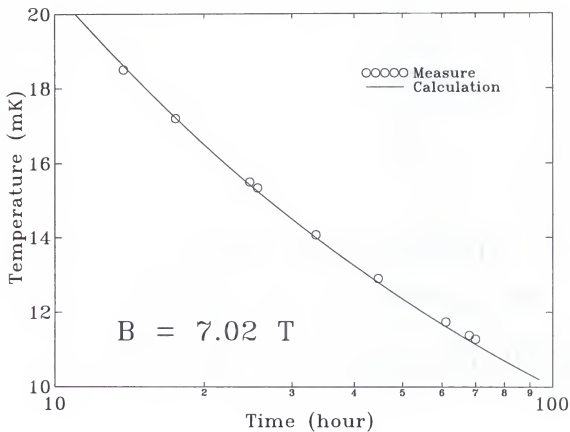


Figure 2.11. Temperature versus time during the precooling of the copper demagnetization stage.

which implies that the cooling (minus sign) of the stage can be expressed as

$$dQ/dt = -\frac{c}{T^2}dT/dt. \quad (2.8)$$

Now, substituting this into Eq. 2.6 and integrating, we have

$$\int_0^t \frac{\frac{k}{2}}{(\frac{k}{2} + a)c} dt' = - \int_{\infty}^T \frac{1}{T'^2} \frac{1}{aT'^2 - b} dT'. \quad (2.9)$$

Performing the integration yields [40]

$$t = \frac{c}{T_0^2} \left(\frac{1}{a} + \frac{2}{k} \right) \left(\frac{1}{2T_0} \ln \frac{T + T_0}{T - T_0} - \frac{1}{T} \right), \quad (2.10)$$

where $T_0 \equiv \sqrt{b/a}$ is the minimum temperature obtained by setting dQ/dt to zero in Eq. 2.4.

The values for the theoretical line in Fig. 2.11 are $k = 130 \text{ mW/K}^2$, $a = 51 \text{ mW/K}^2$, $b = 1.7 \text{ } \mu\text{W}$, and $c = 25 \text{ mJK}$ all of which were obtained from direct measurements or a calculation. In a field of 7.02 T it takes about 72 hours to precooling the copper bundle to 11 mK.

The demagnetization schedule of the copper stage has the following steps:

$$7.02 \text{ T} \xrightarrow{3.5\text{hr.}} 3.74 \text{ T} \xrightarrow{3\text{hr.}} 1.89 \text{ T} \xrightarrow{2\text{hr.}} 0.93 \text{ T} \xrightarrow{2.5\text{hr.}} 0.46 \text{ T} \xrightarrow{2\text{hr.}} 0.23 \text{ T} \xrightarrow{0.5\text{hr.}} 0.19 \text{ T} \xrightarrow{4\text{hr.}} 0.93 \text{ T} \\ \xrightarrow{4\text{hr.}} 0.47 \text{ T} \xrightarrow{4\text{hr.}} 0.23 \text{ T} \xrightarrow{4\text{hr.}} 0.12 \text{ T} \xrightarrow{4\text{hr.}} 0.0058 \text{ T}.$$

This results in the demagnetization profile of the bundle shown in Fig. 2.12. The straight line is for an ideal adiabatic demagnetization given by

$$T = T_i \frac{B}{B_i}, \quad (2.11)$$

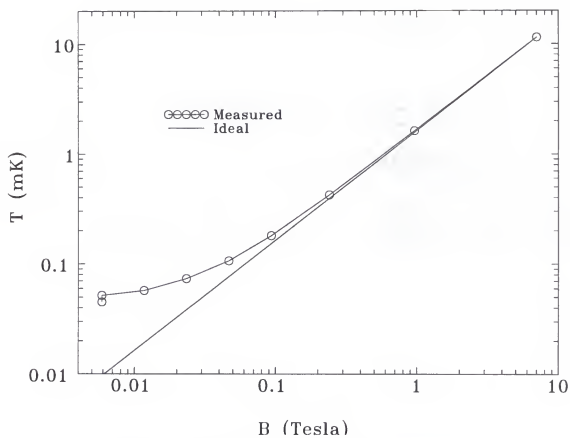


Figure 2.12. Demagnetization curve of the copper stage.

where T_i and B_i are the initial temperature and magnetic field respectively, and B is the current field. The measured curve roughly follows the ideal curve to approximately 1 mK. Below this temperature, the deviation from ideality becomes larger due to parasitic heat capacities and external heat leaks. The minimum measured temperature achieved by this copper stage is 45 μ K.²⁶ Figure 2.12 has two data points in the lowest field. The high temperature point was obtained at the end of the demagnetization process and the low point after closing the fill line of the 1 K pot. Closing the line helps to reduce the vibrational heating and allows the thermometer to reach a lower temperature.

2.5 Thermometry

2.5.1 Melting Pressure Thermometer

The melting pressure thermometer and electronic configuration are similar to those of the experimental cell. However, because it operates in zero field and at a lower pressure, 3.45 MPa (500 psi), the physical properties of the thermometer are slightly different from those of the experimental cell. The strain gauge is made of beryllium copper (BeCu) with a diaphragm of diameter 1.27 cm (0.5") and thickness 0.74 mm (0.029"). The movable plate is epoxied onto a post at the center of the diaphragm, using Stycast 2850FT. The fixed plate is also fixed to the holder with the same epoxy. The plates have a diameter of 1.14 cm (0.45") and are separated by a mylar spacer of thickness 0.05 mm (0.002"). The holders are held together by four 2-56 stainless steel screws.

The body of the thermometer is machined out of a 99.999% pure silver rod with a diameter of 1.3 cm (0.5") and a length of 1.9 cm (0.75"). A coin-silver flange is soldered with eutectic silver-copper solder (71.9% Ag - 28.1% Cu) onto the rod at one end. The

²⁶This temperature was measured by a Pt NMR thermometer located on top of the copper bundle. The temperature at the center of the bundle is assumed to be lower than this value.

flange has eight tapped holes where the strain gauge is bolted. An indium O-ring is used to provide a vacuum seal between the body and the strain gauge. The other end of the rod is welded to a silver plate which has eight clearance holes for screws that are used to attach it to the top of the copper demagnetization stage. Thermal contact between the cell body and the ^3He is provided by silver (Ag) sinter and eight silver rods of diameter 0.8 mm (0.03").

The excitation for capacitance measurements is provided by the internal oscillator of an Ithaco 393 lock-in amplifier. Because the thermometer is operated at melting density, the liquid provides an excellent thermal path to the silver sinter; so the excitation voltage can be kept on at all times without concern about local heating. Also, the stainless steel screws used on the thermometer help to conduct heat away from the ^3He . Note that in the experimental cell, there are no screws nor liquid to remove the heat and it is therefore necessary to turn off the excitation after a measurement to prevent heating of the sample in the experimental cell.

The operating frequency for the thermometer is 1.1 kHz with an amplitude of $1.22 V_{rms}$. A one-to-one isolation transformer is used to keep the system from shorting to ground. The thermometer and its reference capacitor (which sits on the mixing chamber of the dilution refrigerator) are connected to a General Radio 1493 ratio transformer via three coaxial cables as in the experimental cell configuration. The output signal from the bridge is fed into the lock-in amplifier detected, amplified, and then recorded by a chart recorder and a multimeter (HP 3457A) which is connected to a personal computer.

Similar to that of the experimental cell, the strain gauge of the thermometer is trained and calibrated at high temperature. See Sec. 3.1.2 for detailed description of the procedures.

The temperature can be calculated from the pressure using the fitting parameters given in Ref. [9]. For temperature higher than $T_N = 0.934$ mK (the Néel temperature at which the ^3He solid orders into the LFP), the pressure function has the form

$$P(T) - P_N = \sum_{n=-4}^5 A_n T^n, \quad (2.12)$$

where P_N is the pressure measured at T_N , the reference point. The A_n 's are the fitting coefficients and have the following values for P in kPa and T in mK:

$$\begin{aligned} A_{-4} &= 0.78405848 & A_{-3} &= -4.5968629 & A_{-2} &= 9.5436359 \\ A_{-1} &= -10.041752 & A_0 &= 8.5580332 & A_1 &= -4.4431076 \\ A_2 &= 1.5962639 \times 10^{-2} & A_3 &= -3.8815407 \times 10^{-5} & A_4 &= 7.1230765 \times 10^{-8} \\ A_5 &= -6.0945626 \times 10^{-11}. \end{aligned}$$

For temperatures below T_N , the temperature can be determined from the expression

$$P(T) - P_N = A_0 - A_1 T^4, \quad (2.13)$$

with $A_0 = 0.1987$ kPa and $A_1 = 0.2611$ kPa/mK⁴.

2.5.2 Resistance Thermometers

There are total of 17 resistance thermometers at different locations in the cryostat. Six of them are Speer thermometers provided by Oxford, the manufacturer of the dilution unit. The others (R7 – R22) are RuO₂ thermometers [43] which have been calibrated and provided by Professor Mark Meisel. Except for R7 and R8, which are 1 k Ω resistors, the rest are 500 Ω . The resistance values of these thermometers at different temperatures are

listed in Table 2.1 to Table 2.3 as determined by using a Linear Research ac resistance bridge (LR110).²⁷

In this experiment the most important thermometers are R4 and R16 which are located on the mixing chamber. The resistor R4 is used in forming and annealing the high density ^3He samples, procedures which are discussed in the next chapter. The resistor R16 is used to monitor the temperature of the dilution refrigerator after the heat switch has been opened.

²⁷Linear Research, Inc., 5231 Cushman Place, Suite 21, San Diego, CA 92110, phone:(619)299-0719.

Table 2.1. List of the resistance values of the Oxford refrigerator's Speer thermometers at various temperatures.

Labels	R1	R2	R3	R4	R5	R6
Type (Ω)	470	470	100	470	100	220
Locations	Film Burner	Still	Cold Plate	Mix. Chamber	Mix. Chamber	1 K Plate
300 K ($k\Omega$)	0.80	0.80	0.43	0.89	0.46	0.53
77 K ($k\Omega$)	0.86	0.87	0.43	0.98	0.47	0.55
4.2 K ($k\Omega$)	1.20	1.23	0.46	1.42	0.49	0.69

Table 2.2. List of the resistance values of the RuO_2 thermometers above the heat switch at various temperatures.

Labels	R7	R8	R14	R15	R16	R17
Locations	Still Shield	Cold Shield	6 th Heat Exchanger.	7 th Heat Exchanger	Mix. Chamber	Heat Switch Top
300 K ($k\Omega$)	1.31	1.32	0.97	0.97	0.94	0.96
77 K ($k\Omega$)	1.29	1.30	0.94	0.94	0.92	0.94
4.2 K ($k\Omega$)	1.61	1.64	0.89	0.88	0.89	0.91
4 mK($k\Omega$)	—	—	—	—	14.4	19.4

Table 2.3. List of the resistance values of the RuO_2 thermometers below the heat switch at various temperatures.

Labels	R18	R19	R20	R21	R22
Locations	Heat Switch Bottom	Cage Top	Cage Bottom	Bundle Top	Bundle Bottom
300 K ($k\Omega$)	0.96	0.98	0.98	0.98	1.07
77 K ($k\Omega$)	0.94	0.96	0.96	0.96	1.04
4.2 K ($k\Omega$)	0.92	0.93	0.91	0.91	0.92
4 mK($k\Omega$)	18.5	18.8	17.0	27.7	25.2

CHAPTER 3

EXPERIMENTAL PROCEDURES

In this chapter, the calibration procedure of the strain gauges of the experimental cell and melting pressure thermometer is covered in detail, followed by the discussion on the magnetic-field-, ramp-rate-, and temperature-dependence of the experimental cell. A short section on temperature regulation method of the cryostat is given next, proceeded by a detailed description of forming a high-density ^3He sample.

3.1 Calibration of the Strain Gauges

The first half of this section is devoted to the training and calibration of the experimental cell and the second half is for the melting pressure thermometer.

3.1.1 Experimental Cell

In order to translate the capacitance of the strain gauge to the pressure of the sample, the gauge must be accurately calibrated. For high density solid ^3He , the pressure calibration was carried out at 4 K, above the solidification point.

The first step of the calibration process is to train the diaphragm of the gauge to reduce any hysteresis which may exist in the diaphragm. The training is done by slowly pressurizing the cell with ^3He gas from 0.0 to 14.5 MPa (2100 psi) in 15 minutes. This process is carried out by inserting the ^4He -temperature dipstick of the ^3He gas handling system, shown in Fig. 3.1, into a liquid helium-4 (L^4He) dewar. As the stick cools, it cryo-pumps ^3He gas from the storage bottle through the valves N1, #1, and #2, where

"N" denotes a Nupro valve¹ and all other valves are HIP high pressure valves.² The thicker lines represent 0.64 cm (0.25") o.d. Cu tubes and the thinner lines 1.5 mm (0.06") o.d. stainless steel capillaries. The gas is passed through a LN₂ cold trap to remove impurities such as air. When sufficient gas has been removed from the bottle, valve #1 is closed and the dipstick slowly pulled out of the L⁴He dewar increasing the pressure of ³He gas as the stick warms up. With valve #3 opened, the gas pressure can be monitored by a 2000 psi (14 MPa) Heise gauge. Before entering the cryostat through valve #4, the ³He gas is passed through another cold trap to ensure that there is no air contamination. After reaching a pressure of 15 MPa (2100 psi), the gas is slowly removed by inserting the dipstick back into the dewar. When the pressure drops to 7 MPa (1000 psi), the cell is then reloaded with ³He gas at the same rate to 14 MPa (2000 psi). From our experience, this was the most efficient way of training the diaphragm.

Step two of the process is the calibration itself which is carried out by maintaining the cell at different values of pressure and then balancing the capacitance bridge to determine the corresponding capacitance ratio. The calibration is done against the Heise gauge which has a resolution of 7 kPa (1 psi). The calibration range for this cell is between 8 MPa(1200 psi) and 14 MPa (2000 psi). From 14 MPa (2000 psi) the pressure is lowered in 340 kPa (50 psi) steps until it reaches 11 MPa (1600 psi). Then from this pressure down to 10 MPa (1400 psi), the step size is reduced to 70 kPa (10 psi) to give a more accurate calibration in the low temperature working range of the cell. Between 10 MPa (1400 psi) and 8 MPa (1200 psi), the pressure step is increased back to 340 kPa (50 psi). When the minimum pressure of 8 MPa (1200 psi) is reached, the cell is reloaded with ³He gas so

¹Jax Valve & Fitting Company, 3633 Southside Blvd., P. O. Box 16097, Jacksonville, FL 32245, phone: (904) 642-2093.

²High Pressure Equipment Company, 1222 Linden Avenue, Erie, PA 16505-3522, phone: (814) 838-2028, fax: (814)-838-6075.

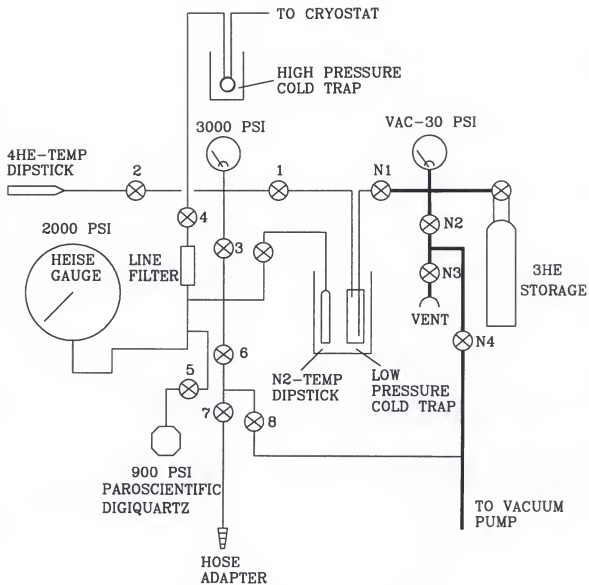


Figure 3.1. ^3He gas handling system.

that the calibration with increasing pressure can be obtained. The pressure steps are the same as in the calibration for decreasing pressure. Upon reaching the maximum pressure of 14 MPa (2000 psi) the gas is again removed in steps for another calibration with decreasing pressure. In a typical calibration, there are two “down” and one “up” pressure calibration processes. By plotting the pressure versus capacitance-ratio-reading for all three processes one can determine the hysteresis of the cell.

Figure 3.2 shows the calibration curves in which circles and triangles denote calibrations with decreasing pressure, and squares the calibration with increasing pressure. A polynomial equation of the form

$$P = a_0 + a_1R + a_2R^2 + a_3R^3, \quad (3.1)$$

was used to fit the down-pressure data points, where P is the value of the pressure, R the corresponding ratio transformer reading, and the a_i 's are fitting parameters. The hysteresis of the cell can be determined by subtracting the fitting polynomial curve from the up pressure data, and this difference is shown in Fig. 3.3. Although this cell has a hysteresis of about 30 kPa (4 psi) for the calibration range between 8 MPa and 14 MPa,³ in practice the hysteresis is much smaller because of a narrower working pressure range of the cell at low temperature. From the temperature at which the sample becomes solid, the total pressure change down to absolute zero is approximately 0.34 MPa (50 psi), 16 times less than the calibration range. Furthermore, during the precooling and demagnetization of the sample the total pressure change is less than 400 Pa making the hysteresis during a measurement much smaller than the 30 kPa measured during the calibration.

³It is possible that the Heise pressure gauge itself has hysteresis.

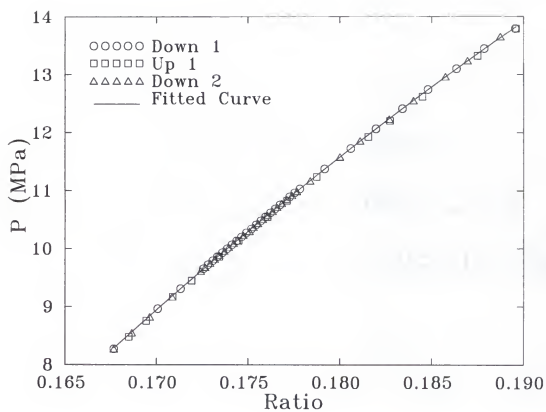


Figure 3.2. Calibration curve of the experimental cell.

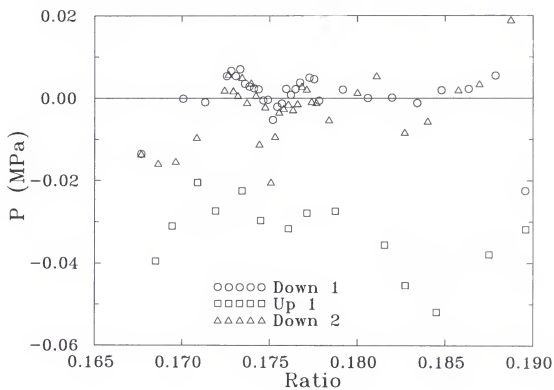


Figure 3.3. Residuals of the calibration curve of the experimental cell.

3.1.2 Melting Pressure Thermometer

Prior to the calibration at around 1 K, the diaphragm is trained. During the process, ^3He is introduced into the thermometer from zero pressure to 3.7 MPa (530 psi) in 5 minutes and then removed at the same rate. When all the gas is out, it is loaded back into the system at a slower rate, about 3.4 kPa/sec (0.5 psi/sec). The pressure is monitored by two pressure gauges, a Heise gauge and a Paroscientific pressure transducer.⁴ The transducer has a sensitivity of 3.4 Pa (0.0005 psi) but can only withstand a maximum pressure of 6.9 MPa (1000 psi) (which makes it unsuitable for calibration of the experimental cell.) When the pressure reaches 3.6 MPa (520 psi), the training is completed.

The calibration range is between 3.6 MPa (520 psi) and 2.8 MPa (400 psi) and is initiated by removing the gas in small pressure steps of 70 kPa (10 psi) down to 3.5 MPa (500 psi). At every step, readings are taken only when the pressure change in 2 seconds is less than 690 Pa (0.01 psi), as determined from the Paroscientific gauge. Between 3.5 MPa (500 psi) and 3.4 MPa (490 psi), the pressure steps are reduced to 7 kPa (1 psi) because this is the operating range of the thermometer at low temperature. Below 3.4 MPa (490 psi), the steps are increased back to 70 kPa (10 psi) until 2.8 MPa (400 psi). Like the calibration of the experimental cell, there are two calibrations with decreasing pressure and one with increasing pressure.

In the next section, we discuss the magnetic-field-, ramp-rate-, and temperature-dependence of the experimental cell. These effects must be subtracted from the raw pressure data to obtain the true pressure change in the sample during the precooling and demagnetization.

⁴Paroscientific, 4500 NE 148 th Avenue Redmond, WA 98052-5126, phone: (206) 883-8700.

3.2 Magnetic-Field-Dependence of the Cell

In our first experimental cell, we encountered a large magnetic-field-dependence which was mainly created by four stainless steel screws used to hold the strain gauge together. Figure 3.4 shows the field-dependence of the first cell expressed as an equivalent pressure change of more than 1.6 kPa between 0 and 1.6 T. This huge field-dependence is about 4 times larger than the expected 400 Pa change in the bcc solid pressure during a demagnetization and made observation of the actual pressure change in the sample difficult. For this reason a second cell was made in an attempt to reduce the effect of the magnetic field.

The second sample cell has no stainless steel screws and exhibits a much smaller field-dependence, as shown in Fig. 3.4. The field-dependence is larger at high fields with a total change of 270 Pa between 1.8 and 2.5 T and the equivalent pressure increases with decreasing field. For fields from 1.8 T down to 0 T, the total change is less than 70 Pa (approximately 4% of that in the first cell) and reverse in direction as shown in Fig. 3.5. Fortunately, the dependence in low fields was reproducible and the pressure change in the sample can be obtained by subtracting the field-dependence from the raw pressure data. The field-dependence was fitted to a polynomial form which was used in analyzing all sample demagnetization data. The fitting form for low fields ($0 \leq B_0 \leq 1.8$ T) is

$$\Delta P = -2.41925 + 67.1808B_0 - 7.15828B_0^2 - 4.3514B_0^3, \quad (3.2)$$

with ΔP in units of Pascals and B_0 in Tesla. The high-field portion ($1.8 < B_0 \leq 2.5$ T) can also be fitted with a polynomial form, though this portion varies from one experimental

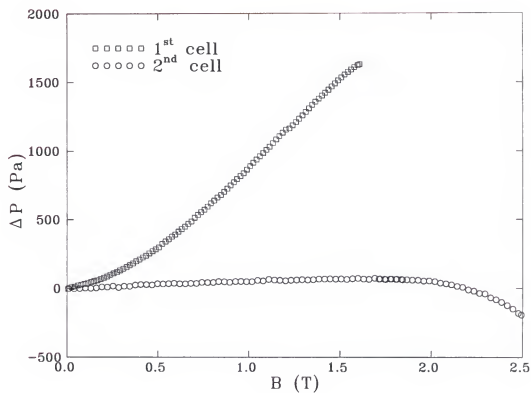


Figure 3.4. Magnetic-field-dependence in decreasing field of the first and second experimental cells at fixed temperature. The first cell has four stainless steel screws and the second cell has none.

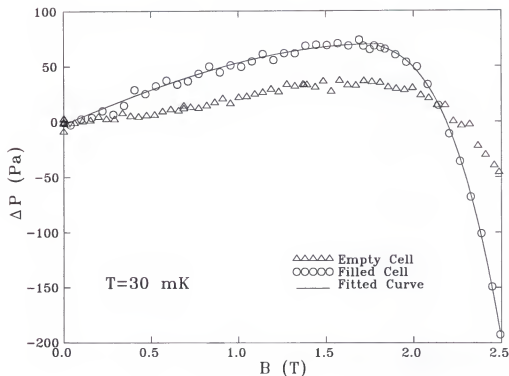


Figure 3.5. Magnetic-field-dependence of the second experimental cell at 30 mK in decreasing fields. The empty-cell curve was measured when there was no sample in the cell while the filled-curve was obtained with a solid sample. A slight difference in the two curves may be due to the different shapes of the diaphragm with and without sample.

run to another. For the filled-cell data in Fig. 3.5, it is given by

$$\Delta P = 2144.74 - 3678.04B_0 + 2188.86B_0^2 - 436.904B_0^3. \quad (3.3)$$

The cell also exhibits a field ramp-rate-dependence as shown in Fig. 3.6. A jump in the equivalent pressure occurred whenever there was a change in the ramp-rate. The equivalent pressure increases when the ramp-rate was decreased and decreases when the rate was increased. The size of the jumps depended on the ramp-rate and the field at which it occurs. The size was smaller in high fields or when the change in the ramp-rates was

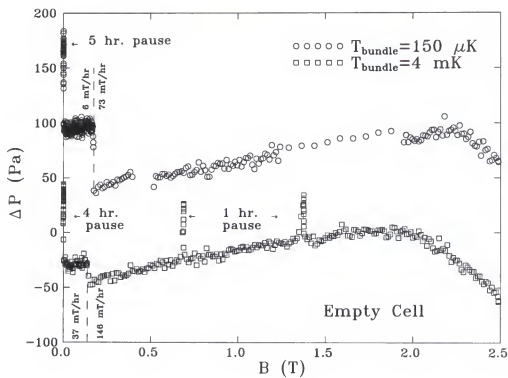


Figure 3.6. Ramp-rate-dependence of the experimental cell at fixed temperatures.

small. The upper curve in Fig 3.6 was obtained at 150 μ K with an initial ramp-rate of -73 mT/hr. At the field of approximately 0.17 T, the ramp-rate was reduced to -6 mT/hr which caused a jump of approximately 60 Pa. When the ramping stopped at zero field, a jump of about 80 Pa was measured during 5 hours of waiting. The bottom curve was obtained at 4 mK with an initial ramp-rate of -146 mT/hr. At the fields of 1.38 and 0.69 T, the ramping was stopped for 1 hour and two equivalent pressure increases of 40 and 50 Pa, respectively, were observed. At the field of 0.14 T, the ramp-rate was reduced to -37 mT/hr which caused a jump of approximately 20 Pa. When the ramping was stopped at zero field, the equivalent pressure increased by 70 Pa over 4 hours of waiting. The measurements of the ramp-dependence were made in an empty cell, but this effect could still be seen in the filled cell. In order to avoid this dependence during a sample demagnetization, the demagnetization rate was kept constant at all times. Since there existed a large equivalent pressure jump at zero field, all data taken after the demagnetization were not included in the data analysis.

We do not know the cause of this ramping effect but believe that it is not due to eddy current heating.⁵ In Fig. 3.7 we have plotted the square of the ramp-rate versus the difference in the squares of the temperatures of the capacitance plates and those of the copper bundle. The temperature of the plates was determined by calculating the size of the pressure jumps and relating these values to the temperature-dependence (discussed in the next section) of the cell. If the ramping effect were caused by the eddy-current heating, the fitted curve would have $\dot{B}^2$ proportional to $T_{plate}^2 - T_{bundle}^2$, but we find no such dependence.

⁵An induced magnetic force on the diaphragm can also be discounted because this force would cause an increase in the equivalent pressure with a magnitude of only 10^{-4} to what observed.

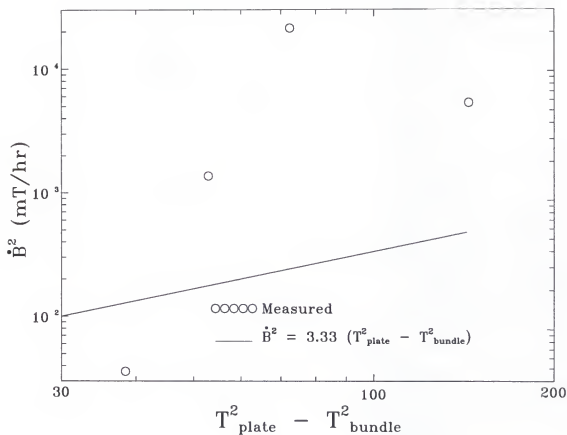


Figure 3.7. Ramp-rate versus temperature. The fit line shows that the heating, if real, is not from the metal plates.

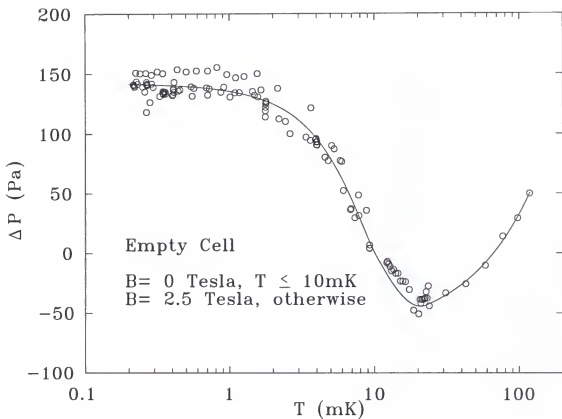


Figure 3.8. Temperature-dependence of the experimental cell when it is empty.

3.3 Temperature-Dependence

Besides the field and ramp-rate-dependence, the cell also has a small temperature-dependence. The temperature-dependence was measured for an empty cell⁶ and is shown in Fig. 3.8. For temperature below 10 mK, data were taken in only zero field because of the long thermal-relaxation time of the cell in high fields. The temperature-dependence was fitted to the polynomial form $P = a_0 + a_1T + a_2T^2 + a_3T^3$ in three temperature regions, $T \leq 10$ mK, $10 < T \leq 23$ mK, and $T > 23$ mK. In this work the region of $T \leq 10$ mK was the most important because most of the pressure change during a sample precooling occurred

⁶The temperature-dependence of a filled cell could not be measured because the sample has its own temperature-dependence.

Table 3.1. Fitting coefficients of the temperature-dependence of the experimental cell. The temperature-dependence has the polynomial form $P = a_0 + a_1T + a_2T^2 + a_3T^3$.

	a_0	a_1	a_2	a_3
$T \leq 10$ mK	142.689	-5.15486	-2.12073	0.121375
$10 < T \leq 23$ mK	170.696	-25.6000	0.971897	-1.14433×10^{-2}
$T > 23$ mK	-66.4553	1.01595	-2.13444×10^{-4}	0

at these temperatures. The small temperature-dependence below 1 mK was not significant during the demagnetization of the sample.

3.4 Temperature-Regulation Method

The temperature of the mixing chamber was monitored and regulated using the arrangement shown in Fig. 3.9. The components involved are a Linear Research 110 (LR-110) resistance bridge, a Speer 400 resistance thermometer (R_4) located on the mixing chamber, an Oxford 1 k Ω heater and a LR-130 temperature controller. The value of R_4 was measured by the LR-110 resistance bridge whose DC error output was used as feed back to the temperature controller. The LR-130 processed the DC signal and delivered an appropriate amount of current to the heater to stabilize the value of R_4 . For this cryostat it was found that the best settings for the LR-110 resistance bridge were: the excitation voltage $V_{exc} = 0.3$ mV_{rms}; fine=100%, and the time constant $\tau = 10$ seconds. For the LR-130 temperature controller, the settings were: DC gain = 30; variable integration time constant $T_1 = 30$ seconds; high frequency roll-off time constant $T_2 = 0.03$ seconds; variable derivative time constant $T_3 = 10$ seconds; and output resistance $R_{out} = 1$ k Ω with the monitor port shorted to bypass the 1 k Ω current-sensing resistor.

3.5 Forming the Samples

All three samples, 20.10, 19.65 and 19.61 cm³/mole, used the same ³He gas which had approximately 14 ppm of ⁴He impurity. The impurity value was measured using a Veeco

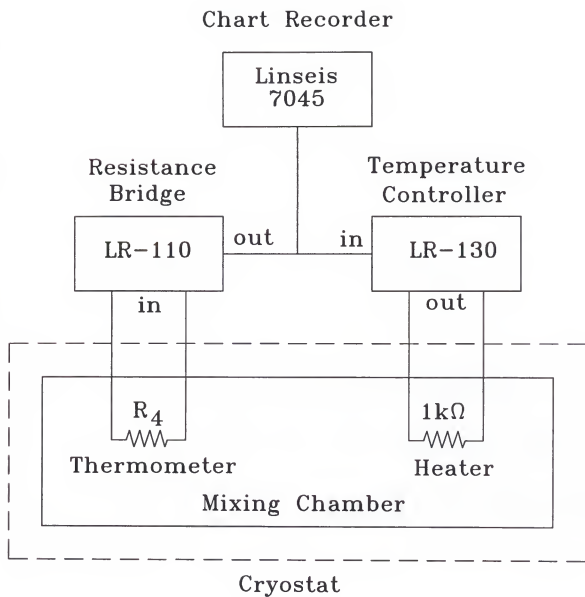


Figure 3.9. Schematic drawing of the temperature-control system.

MS-17 leak detector⁷ which had been calibrated with a sample of ^3He and ^4He gas of known ratio. The blocked-capillary technique was used in forming the samples. To grow a sample at a desired density, one must first determine at what pressure the sample would begin to freeze. For example, if the desired molar volume was $19.61 \text{ cm}^3/\text{mole}$ then the sample would begin to freeze near 14 MPa ($\approx 2000 \text{ psi}$) indicated by point b' in Fig. 3.10. Because the temperature was lower near the 1 K pot, a solid block was likely to form in the fill line before the bulk sample could reach the melting curve. If this happened, one could not supply gas to the cell to maintain the necessary pressure and the sample would reach the melting curve at a lower pressure, thus yielding a lower density.⁸ It was therefore necessary to load the cell with a pressure slightly higher than the targeted melting pressure. The loading pressure of our hcp samples was about 14.1 MPa (2050 psi). Since the solidification temperature of high-density solid ^3He near the bcc-hcp phase boundary is approximately 3.2 K, to form a high-quality sample requires good control of the temperature in this range. Below is the description of how high stability in temperature above 1 K was achieved in this cryostat.

First, the $^3\text{He}/^4\text{He}$ mixture was removed from the dilution refrigerator.⁹ Second, the 1 K pot was filled and pumped and used as the cooling source in the cryostat. When cooling the pot to 1 K, it is pumped on by the Kinney pump through the wide open #45 valve, shown in Fig. 2.8. In order to maintain higher temperatures, the pumping rate must be controlled using valve #47 and #50, with valve #48 used as a throttle. For example, to

⁷Veeco Instruments, Inc., Terminal Drive, Plainview, NY 11803, phone: (516) 349-8300

⁸On this cryostat, one can melt the solid block in the fill line by applying 60 mW (10 mA) to the still heater.

⁹Especially the ^3He gas left inside the refrigerator would condense in the heat exchangers and drip into the mixing chamber causing large temperature fluctuations.

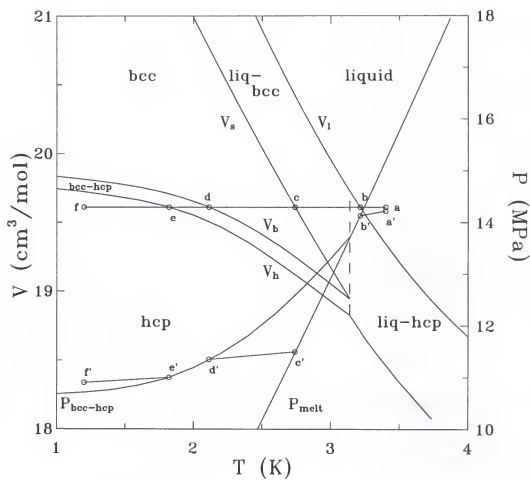


Figure 3.10. Molar volume and pressure versus temperature for high density solid ^3He . The line a-f represents a molar volume of $19.61 \text{ cm}^3/\text{mole}$ with the corresponding pressure change in the sample is indicated by the path a'-f'.

achieve a temperature of 3.2 K indicated by point b, b' in Fig. 3.10, the throttle valve was opened wide enough so that the average pressure of the pot was about 32 kPa [44].¹⁰

Third, to cool other parts a small amount of ⁴He gas was circulated in the dilution refrigerator using the Roots and Alcatel pumps. The gas was that remaining in the back of the Alcatel pump, at a pressure of 500 mTorr, after all the mixture was removed from the refrigerator and stored in the storage tanks.

When the temperature was regulated properly (see Section 3.4, the refrigerator was left to cool overnight to 3.4 K, point a, a' in Fig. 3.10, a temperature slightly above the solidification point b, b' of the sample. For our two hcp samples, 19.61 and 19.65 cm³/mole, point b, b' was about 3.2 K corresponding to a value of 1540 Ω of R₄.

After the pressure in the cell was set, the refrigerator was cooled by decreasing the regulated temperature in 0.1 K (100 Ω on R₄) steps. Along the melting curve, the temperature was held fixed for 5 minutes at three different values which were about 0.02 K ($\Delta R_4 = 20 \Omega$) apart, to ensure uniform sample density. Figure 3.11 shows the pressure decrease during the formation of the 19.61 cm³/mole sample.¹¹

After the sample was completely solidified, it was annealed for 3 days (just below point c, c') until the pressure decrease became negligible. Figure 3.12 shows the pressure change versus time during annealing. During the first 5 hours the pressure increased by approximately 2 kPa (0.3 psi), then began to decrease. For the 65 hours of annealing, the total pressure decrease was approximately 0.16 MPa (23 psi). The sample was then cooled slowly by decreasing the temperature in steps with 10-minute waiting time per step. The size of the first 9 steps was only 0.1 mK (1 Ω), to minimize the thermal shock to the sample,

¹⁰With these settings, the pressure of the pot fluctuated between 17 kPa (2.8 K) and 47 kPa (3.5 K) with a period of approximately 1 hour.

¹¹The three spikes on the melting curve are due to a slight overshoot on the temperature controller.

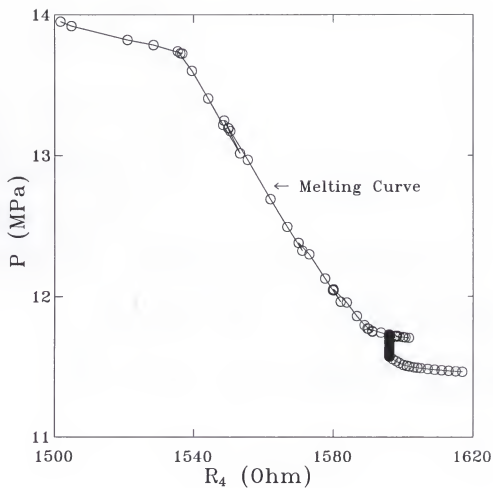


Figure 3.11. Pressure versus the resistance thermometer during the formation of the $19.61 \text{ cm}^3/\text{mole}$ sample. The pressure drop at 1596Ω occurs during the annealing of the sample.

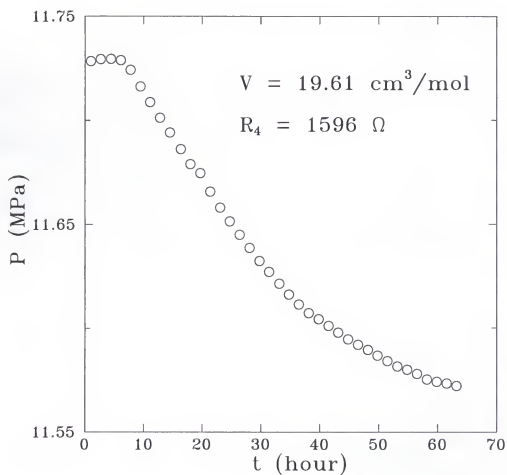


Figure 3.12. Pressure versus time during annealing near the melting curve of the $19.61 \text{ cm}^3/\text{mole}$ sample.

following by six 0.2 mK ($2\ \Omega$) steps. Then the step-size was increased further to 0.3, 0.4, 0.5, 1, and 2 mK with three steps each. The last 2 mK step concluded the sample annealing process for a bcc sample; however, a low-density hcp solid requires another annealing which occurs after the sample crosses the bcc-hcp phase boundary (see Fig. 3.10).

The bcc-hcp transition was marked by a sharp drop in pressure. In our hcp sample, the drop was measured to be approximately 90 kPa (13 psi), see Fig. 3.13. Additional drops in pressure were achieved by changing the temperature of the sample back and forth below the bcc-hcp transition which was the procedure used for the $19.65\ \text{cm}^3/\text{mole}$ hcp sample. Alternatively, for the $19.61\ \text{cm}^3/\text{mole}$ sample, the same pressure drop was achieved by lowering the temperature slowly over a period of a week. The pressure drop at a fixed temperature below the bcc-hcp transition is shown in Fig. 3.14. The annealing data indicate that the solid conversion from bcc to hcp takes place in a wide temperature range below the initial transition. In Fig. 3.13, the pressure change can be seen occurring with R_4 as large as $1900\ \Omega$, about 150 mK below the bcc-hcp phase line. However, despite the fact that there was additional pressure drop during the sample annealing, the total measured pressure decrease was only 160 kPa which was 200 kPa smaller than what expected in a large bulk solid [45].¹² This phenomena may relate to the small thickness of the bulk sample, which was only 0.13 mm. The smaller pressure decrease at the bcc-hcp transition was also reported by Okamoto [27], whose sample thickness was 0.15 mm. For a sample of molar volume of $19.66\ \text{cm}^3/\text{mole}$, he found a total pressure decrease of only 60 kPa which was 250 kPa smaller than the expected value for a bulk sample [45].

¹²The effect of a non-constant sample volume can only account for approximately 24 kPa of the missing pressure drop.

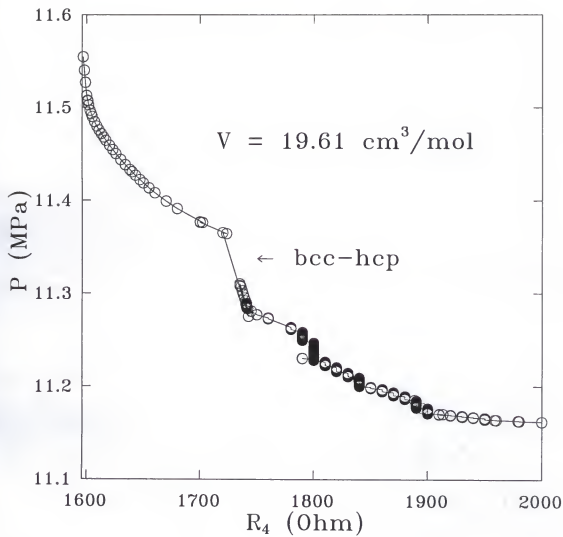


Figure 3.13. Pressure versus resistance thermometer for the $19.61 \text{ cm}^3/\text{mole}$ sample around the bcc-hcp phase line.

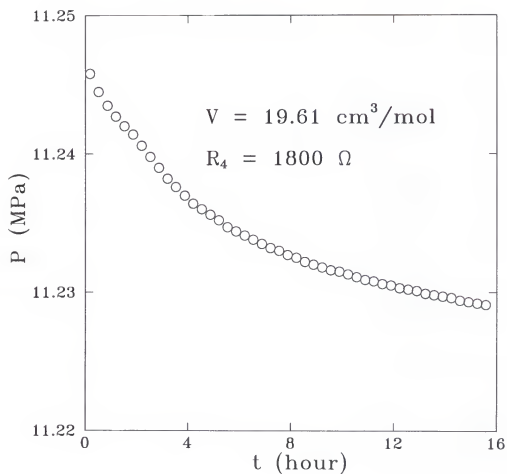


Figure 3.14. Pressure change at a fixed temperature during the annealing of the $19.61 \text{ cm}^3/\text{mole}$ sample below the bcc-hcp phase line.

CHAPTER 4

EXPERIMENTAL RESULTS

In this chapter, the experimental results are presented with the precooling and demagnetization data shown for a bcc sample of molar volume $20.10 \text{ cm}^3/\text{mole}$ and two hcp samples with molar volumes of 19.61 and $19.65 \text{ cm}^3/\text{mole}$. The results are then discussed in relation to predictions of theoretical models.

4.1 bcc Sample

A bcc sample with a molar volume of $20.10 \text{ cm}^3/\text{mole}$ was used as a test to evaluate our experimental procedure prior to our measurements on hcp samples. A bcc sample is useful for this purpose because most of its magnetic characteristics have been determined at lower density. In addition, the pressure has been measured during demagnetization of a bcc solid sample [6] providing excellent data for comparison.

4.1.1 Precooling Data

Figure 4.1 shows the entire cooling process of a ^3He sample. Our bcc sample was precooled in a field of 2.5 Tesla over a period of 40 days to a temperature below 0.55 mK, thereby removing 99% of its entropy, by demagnetizing the copper stage. The precooling process was long due to the large Kapitza resistance [35] between the ^3He solid and the silver sinter. For magnetic fields higher than 0.2 T, the resistance has a $1/T^2$ dependence which gives rise to a longer thermal relaxation time of the sample as the temperature decreases.

Table 4.1. Precooling data for $v = 20.10 \text{ cm}^3/\text{mole}$ bcc solid in 2.5 T field. T_{bundle} is the copper bundle temperature measured by the melting pressure thermometer, p^2 is the square of the polarization, ΔP is the total pressure change of the sample from high temperature, and the waiting time is the amount of time during which T_{bundle} was held constant.

$T_{\text{bundle}}(\text{mK})$	p^2	$\Delta P \text{ (Pa)}$	Waiting time(hour)
6.19	0.091	-18.6 ± 2.4	19
3.2	0.29	-43 ± 7.7	26
2.55	0.408	-131.7 ± 19.1	115
1.75	0.642	-129.6 ± 7.2	44
0.88	0.952	-317.1 ± 8.9	121
0.55	0.990	-284 ± 1.1	235

However, this large resistance is beneficial in preventing a large amount of heat flow from the cell into the sample during the demagnetization of the sample.

The precooling of the sample was carried out in steps. First, the sample was precooled for 107 hours¹ along with the copper bundle to a temperature near 11 mK. Second, the copper bundle was demagnetized to 6 mK where the temperature was held constant for 19 hours allowing the sample to cool to the new temperature. Then the temperature of the copper bundle was reduced further, with this process continuing until the sample reached 0.55 mK as shown in Table 4.1. At each temperature, the pressure change ΔP was determined by fitting to the formula:

$$\Delta P = P_0 + P_1 \exp(-t/\tau) \quad (4.1)$$

where $P_0 + P_1$ is the pressure at $t = 0$ when the bundle reaches the new temperature, and τ is the thermal time constant.

The pressure change in the sample versus the square of the polarization which is given by $p = \tanh(\gamma \hbar B_0 / 2k_B T)$ and the temperature are plotted in Figs. 4.2 and 4.3, respectively.

¹A typical precooling takes about 72 hours; however, in this run our dilution refrigerator was partially blocked and the circulation rate was reduced to maintain an operational return-line pressure.

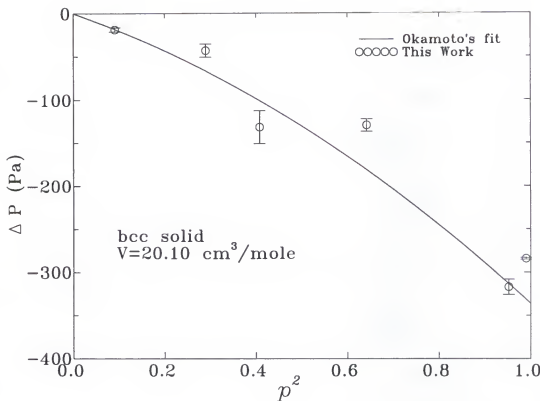


Figure 4.2. Pressure change versus the square of the polarization during the bcc sample precooling.

The solid lines in both graphs were obtained from Okamoto's work on a bcc sample with a molar volume of $19.99 \text{ cm}^3/\text{mole}$ [6] scaled to the molar volume of our sample. While our objective was not to determine the precooling with high accuracy, our precooling data agree well with those of the Tokyo group. Using the data in Fig. 4.2 together with Eq. 1.35, one can determine the volume derivatives, $\partial\theta_w/\partial v$ and $\partial K_p/\partial v$ for a bcc solid. For their bcc solid, Okamoto *et al.* [6] found $\partial\theta_w/\partial v = -40.7 \pm 2.2 \text{ } \mu\text{K mole}/\text{cm}^3$ and $\partial K_p'/\partial v = -11.0 \pm 0.7 \text{ } \mu\text{K mole}/\text{cm}^3$.

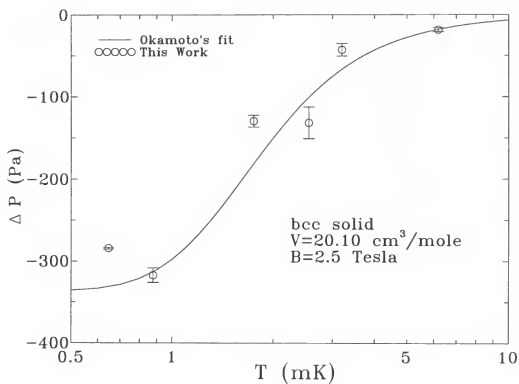


Figure 4.3. Pressure change versus temperature during the bcc sample precooling.

4.1.2 Demagnetization Data

After precooling in 2.5 T to 0.55 mK, the sample was demagnetized with a ramp-rate of -73 mT/hour. This rate was kept constant to zero field in order to avoid the spikes that occur with changes in ramp-rate (see Fig. 3.6). Figure 4.4 shows the pressure curve of the bcc sample during its demagnetization, with the field-dependence of the cell subtracted. A similar pressure behavior was observed by Okamoto *et al* [6]. The pressure remained flat until 0.5 T and then began to rise, indicating that the sample had entered the high-field phase from the paramagnetic phase. The pressure increased rapidly between the fields of 0.5 T and 0.02 T with a total change of 340 Pa. A second transition, marked by a pressure jump of 20 Pa, occurred at 0.02 T when the sample entered the LFP from the HFP. The observation of this transition indicates that our bcc sample was cooled to a temperature below 30 μ K. The pressure of our sample remained flat in the LFP to zero field, unlike the Tokyo data [6] which exhibited a pressure decrease of 35 Pa in the LFP. The absence of a pressure drop at low fields indicates that the use of a StyCAST seal instead of an indium O-ring in our sample cell significantly reduced heating near the end of the sample demagnetization. This knowledge is important in explaining the results of our hcp samples.

4.1.3 Discussion of bcc Results

To understand better the bcc pressure data, it is useful to examine a few adiabatic demagnetization paths on the $B - T$ phase diagram for different entropies. For each of these paths, the value of the entropy was determined from the initial temperature and field at the beginning of the demagnetization. The $B - T$ phase diagram was constructed for the bcc sample using the results from two previous experiments. The LFP-PP, LFP-HFP

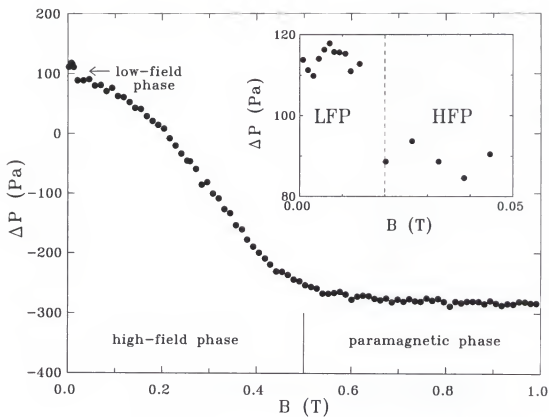


Figure 4.4. Pressure change during the bcc sample demagnetization with nearly 100% polarization.

and low-field PP-HFP boundaries were scaled from the work of Xia *et al.* [13] at melting density using scaling factors of v^{18} for temperature and $v^{14.5}$ for field [12]. The high-field PP-HFP boundary was scaled from work of Fukuyama *et al.* [46] in which the magnetic phase diagram of the bcc solid at a molar volume of $22.69 \text{ cm}^3/\text{mole}$ was determined by pressure measurements. The scaling factor used for both the temperature and the fields was v^{18} .

Adiabatic demagnetization curves were generated and scaled to our sample using the constant-field entropy data at melting density published by Ni *et al.* [29]. The scaling factors were v^{18} and $v^{14.5}$ for the temperatures and the fields, respectively. Figure 4.5 shows three predicted paths on a $B-T$ phase diagram for adiabatic sample demagnetizations of the bcc solid with $S/R\ln 2 = 0.1, 0.3$, and 0.7 . The entropy values can be determined by substituting in the initial values of the temperature and applied field into the equation:

$$S = R(\ln(2 \cosh x) - x \tanh x), \quad (4.2)$$

where R is the gas constant; x is a dimensionless parameter given by $\gamma \hbar B_0 / 2k_B T$ with B_0 the applied field.²

For $S = 0.1R\ln 2$ the sample enters the HFP at about 0.3 T , with the cooling rate decreasing with decreasing field. The HFP-LFP boundary is reached near 0.03 T and $21 \text{ } \mu\text{K}$. Then, because of the lower entropy in the LFP, the sample warms slightly along this boundary to a temperature of $26 \text{ } \mu\text{K}$ where it enters the LFP and remains to zero field. The demagnetization path of our bcc sample was similar to that for $S = 0.1R\ln 2$, except that our sample entered the HFP at slightly higher field because its remaining entropy

²The above equation is only valid in high magnetic fields where the Zeeman energy is dominant.

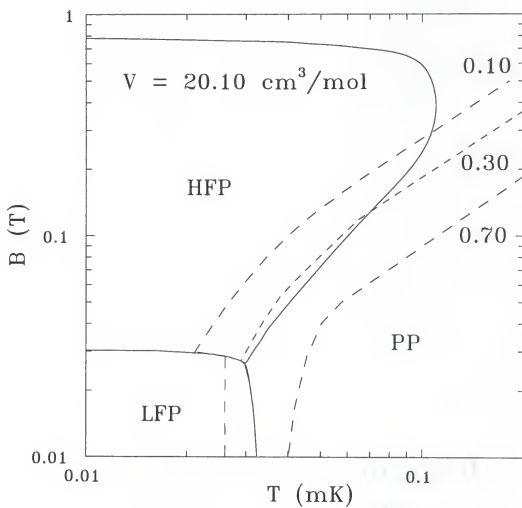


Figure 4.5. Constant-entropy paths during a bcc sample demagnetization.

was almost 0. There is also a slight disagreement in the value of B_{c1} (HFP-LFP). In our measurement, we found B_{c1} to be about 0.02 T yielding a scaling factor of 16.7 which is higher than the 14.5 ± 0.3 obtained in Fukuyama's work [12]. For their bcc sample with a molar volume of $19.99 \text{ cm}^3/\text{mole}$, Okamoto *et al.* [6] reported B_{c1} to be 0.027 T which yielded a scaling factor of about 14.7. However, because the applied magnetic field was trapped when the indium O-ring became superconducting ($B_c = 0.029 \text{ T}$), Okamoto and co-workers were forced to measure the trapped field in a dummy cell and then assumed that the same field was trapped in the real cell in order to deduce the value of the field at the sample. In our experiment, the trapped-field problem was totally avoided because our cell has no indium O-ring and the value of the field was measured directly. Therefore, the measured value for B_{c1} in our sample is more reliable than that of Okamoto *et al.* who might have over-estimated the value of the trapped field in their cell. The question of the scaling factor for $B_{c1}(v)$ bears further investigation because of its implication for the volume-dependence of various exchange parameters.

The second demagnetization curve is for $S = 0.3R\ln 2$. In the HFP, the behavior is almost like that for $S = 0.1R\ln 2$; however, the sample never enters the LFP fully but follows the HFP-LFP and then the PP-LFP phase lines to zero field. In other words, the sample is only partially ordered at low fields. The third curve represents the demagnetization curve of a sample with $S = 0.7R\ln 2$. The sample neither enters the HFP nor the LFP, remains in the PP even at zero field.

The predicted path for $S = 0.1R\ln 2$ is qualitatively in agreement with our bcc experimental data. Both results clearly show two magnetic ordering transitions (PP-HFP and HFP-LFP) and the observation of the HFP-LFP transition indicates that the sample was cooled below $30 \text{ } \mu\text{K}$. The predicted paths also show that magnetic ordering can be achieved

only at a certain value of entropy, and different initial conditions will yield different results. This knowledge is extremely useful in examining the pressure data of hcp samples.

4.2 hcp Sample

In two separate coolings from room temperature in March 1995 and October 1995, pressure measurements were made on two hcp samples with molar volumes of $19.65 \text{ cm}^3/\text{mole}$ and $19.61 \text{ cm}^3/\text{mole}$, respectively. This section is divided into two parts, one for the precooling data and the other for the demagnetization data. There were a total of seven precoolings and demagnetizations, three for the first sample and four for the second, with conditions as described in the following text.

4.2.1 Precooling Data

In the first precooling, the hcp sample ($v = 19.65 \text{ cm}^3/\text{mole}$) was cooled to a temperature of 0.61 mK while in a field of 2.5 T . The duration of the entire process was 38 days and at the end the spins were almost fully polarized with about 98% of the spin entropy removed from the sample.

As in the precooling of the bcc sample, the hcp precooling was carried out by slowly stepping down the temperature of the copper bundle. In the first step, the sample was precooled by the dilution refrigerator, along with the copper bundle, to 11.2 mK in 84 hours. The temperature of the bundle was then lowered further by demagnetization as shown in Table 4.2. As discussed in the previous section for the bcc sample, the pressure change of the hcp sample for each precooling steps was fitted to an exponential form. Figure 4.6 shows the plot of ΔP versus p^2 of the sample during three different precoolings. Here, p^2 was determined by the applied field B_0 and the precooling temperature T using

Table 4.2. First precooling data for $v = 19.65 \text{ cm}^3/\text{mole}$ hcp solid in 2.5 T field. T_{bundle} is the temperature of copper bundle measured by the melting pressure thermometer, p^2 is the square of the polarization of the spins, ΔP is the total pressure change of the sample from high temperature, and the waiting time is the amount of time during which T_{bundle} is held constant.

$T_{\text{bundle}}(\text{mK})$	p^2	ΔP (Pa)	Waiting time(hour)
6.06	0.095	13.1 ± 1.6	24
3.38	0.266	24.6 ± 4.6	85
2.45	0.431	55.5 ± 13.5	71
1.78	0.631	36.1 ± 19.3	22
0.954	0.932	147.5 ± 10.3	142
0.614	0.993	119.6 ± 0.8	92

Table 4.3. Second precooling data for $v = 19.65 \text{ cm}^3/\text{mole}$ hcp solid in 2.5 T field.

$T_{\text{bundle}}(\text{mK})$	p^2	ΔP (Pa)	Waiting time(hour)
6.73	0.078	10.8 ± 9.5	19
3.61	0.239	18.3 ± 4.6	64
2.79	0.358	22.1 ± 21.3	21

the equation $p = \tanh(\gamma \hbar B_0 / 2k_B T)$. The circles represent the first precooling data and the solid line is the fitted curve extrapolated from the Tokyo data [6] for this sample.

In the second and third precoolings, the sample was cooled to a temperature near 3 mK in a 2.5-T field. The amount of entropy remaining in the system was approximately $0.75R \ln 2$ with a polarization of 50%. The reason for leaving the higher entropy in the sample is discussed in Section 4.2.2. The two precooling sequences are given in Tables 4.3 and 4.4.

For the second hcp sample with $v = 19.61 \text{ cm}^3/\text{mole}$, there were four precoolings (the fourth, fifth, sixth, and seventh of the precoolings of hcp samples). In the fourth precooling,

Table 4.4. Third precooling data for $v = 19.65 \text{ cm}^3/\text{mole}$ hcp solid in 2.5 T field.

$T_{\text{bundle}}(\text{mK})$	p^2	ΔP (Pa)	Waiting time(hour)
7.02	0.072	10.0 ± 9.6	19
3.90	0.209	45.6 ± 4.7	96
3.00	0.321	35.1 ± 25.1	19

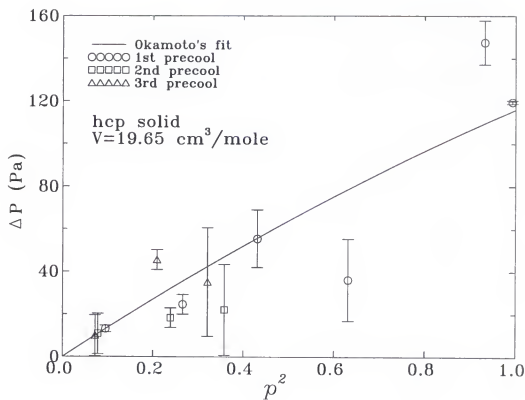


Figure 4.6. Pressure change versus the square of the polarization during the sample pre-cooling of the first hcp sample.

Table 4.5. Fourth precooling data for $v = 19.61\text{cm}^3/\text{mole}$ hcp solid in 2.5 T field.

$T_{\text{bundle}}(\text{mK})$	p^2	ΔP (Pa)	Waiting time(hour)
6.13	0.093	12.9 ± 1.1	46
3.99	0.201	39.1 ± 4.0	90

Table 4.6. Fifth precooling data for $v = 19.61\text{cm}^3/\text{mole}$ hcp solid in 2.5 T field.

$T_{\text{bundle}}(\text{mK})$	p^2	ΔP (Pa)	Waiting time(hour)
6.05	0.095	13.1 ± 1.2	65
3.48	0.254	23.9 ± 1.6	116
1.77	0.635	58.1 ± 0.8	325

the sample was cooled to 4 mK with a remaining entropy of $0.85R\ln 2$. In the fifth precooling, the sample had a final temperature of 1.77 mK corresponding to a remaining entropy of $0.46R\ln 2$. The sixth and seventh precoolings had the same final temperature of 1.20 mK corresponding to $0.23R\ln 2$ entropy. The seventh precooling was a repetition of the sixth because of a large heat leak encountered during the sixth demagnetization of the hcp sample. Tables 4.5, 4.6, and 4.7 list the sequences for the fourth, fifth, and sixth precoolings of the hcp sample, respectively. Figure 4.7 shows a plot of ΔP versus p^2 for the second sample with three different symbols representing the three different precoolings.

4.2.2 Demagnetization Data

In this section, the first observation of magnetic ordering of a hcp sample is presented. Our results indicate that the pressure of the sample behaves differently depending on the amount of entropy removed during the precooling. By comparing our data for different entropies to calculations for the high-temperature expansion of the multiple-exchange and

Table 4.7. Sixth precooling data for $v = 19.61\text{cm}^3/\text{mole}$ hcp solid in 2.5 T field.

$T_{\text{bundle}}(\text{mK})$	p^2	ΔP (Pa)	Waiting time(hour)
6.36	0.087	11.1 ± 1.0	41
1.74	0.645	91.1 ± 23.5	114
1.20	0.851	83.1 ± 0.7	472

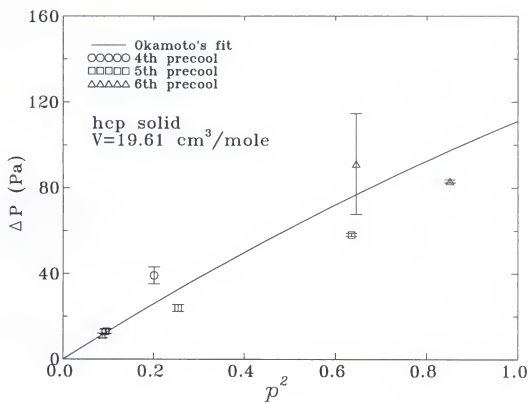


Figure 4.7. Pressure change versus the square of the polarization during the sample pre-cooling of the second hcp sample.

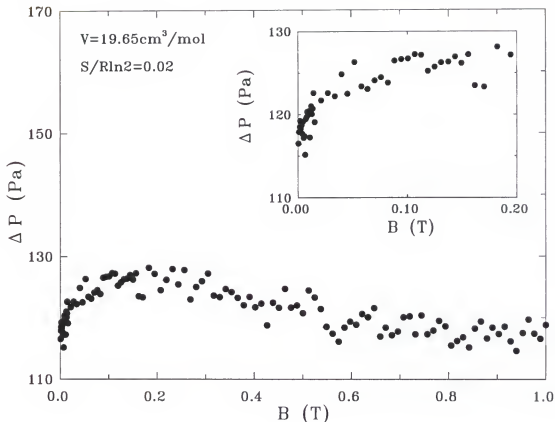


Figure 4.8. Demagnetization data for the $v = 19.65 \text{ cm}^3/\text{mole}$ sample with entropy of $0.02R\ln 2$.

Heisenberg models discussed in Section 4.2.3, we have strong evidence that our low-entropy hcp sample was magnetically ordered at low temperature. The demagnetization data for the first sample, $v = 19.65 \text{ cm}^3/\text{mole}$, is discussed first followed by a discussion of the results from the second sample, $v = 19.61 \text{ cm}^3/\text{mole}$.

The first demagnetization of the hcp samples ($v = 19.65 \text{ cm}^3/\text{mole}$) started with 98% entropy removal. The change in pressure versus field during the demagnetization is shown in Fig. 4.8. The pressure increased by approximately 6 Pa over the field range from 1.8 T to 0.2 T and then began to drop, with a total amount of 10 Pa between 0.2 T and 0 T. A similar pressure decrease in low fields was also observed by the Tokyo group [6]; however,

they attributed this pressure drop to heating of their hcp samples. In our case no heating occurs at low fields as demonstrated for the bcc solid.

In a previous publication [47], we raised the possibility that the decrease in pressure was the signature of ordering of the hcp solid. To demonstrate that this is the case, we carried out two tasks. First, we made pressure measurements on samples with high entropies so that the samples could not magnetically order near the end of a demagnetization. These results showed the behavior of unordered samples during a sample demagnetization and allowed us to make a comparison with the results for a low-entropy sample in which magnetic ordering occurred. Second, we used theoretical models for comparison with our pressure measurements on high-entropy samples. For the second task, we employed the high-temperature expansion of the multiple-exchange and Heisenberg models, described in Section 4.2.3. The pressure measurements of high-entropy samples are discussed below.

The second precooling ended when the sample ($v = 19.65 \text{ cm}^3/\text{mole}$) reached a temperature of approximately 2.8 mK, corresponding to a remaining entropy of $0.72R\ln 2$. During the sample demagnetization, shown in Fig. 4.9, the pressure increased by approximately 11 Pa between 1.8 and 0.1 T, a behavior quite similar to that of the first precooling in which $S \approx 0$. However, below 0.1 T, the pressure increased rapidly with a total change of 26 Pa in going to zero field (from ~ 34 Pa at 0.1 T to ~ 60 Pa at 0 T). The increase in pressure was quite a contrast to the behavior observed in the first sample demagnetization. The implication of this different behavior will be discussed in Section 4.2.3.

Our third hcp-sample demagnetization was with $0.75R\ln 2$ remaining entropy and we observed a similar pressure increase. The only major difference between the second and the third demagnetization was the temperature of the copper bundle, which was kept at 0.5 mK near the end of the third demagnetization instead of 0.1 mK for the second demagnetization.

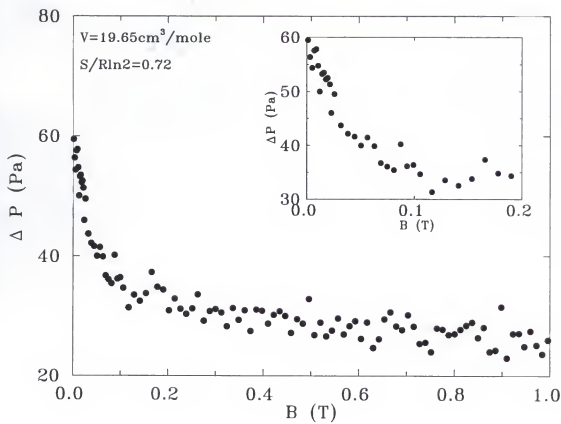


Figure 4.9. Demagnetization data for the $v = 19.65 \text{ cm}^3/\text{mole}$ sample with entropy of $0.72R \ln 2$.

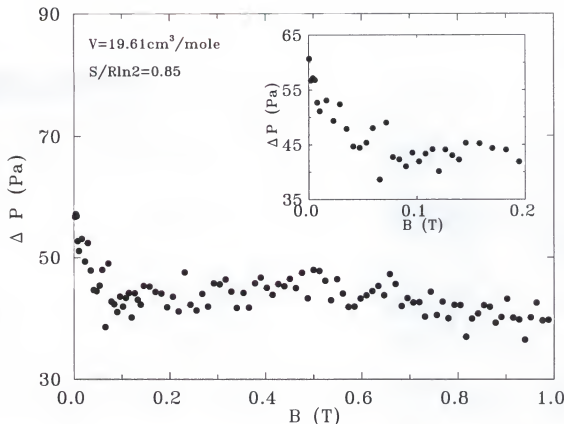


Figure 4.10. Demagnetization data for the $v = 19.61 \text{ cm}^3/\text{mole}$ sample with entropy of $0.85R\ln 2$.

The final bundle temperature in the first demagnetization was also 0.5 mK and we wanted to be sure that the pressure increase that we first encountered in the second demagnetization was not related to the temperature of the copper bundle.

The fourth sample demagnetization ($v = 19.61 \text{ cm}^3/\text{mole}$) was with $S = 0.85R\ln 2$. As shown in Fig. 4.10, the pressure of the sample rose approximately 20 Pa towards the end of the demagnetization. As will be discussed in Section 4.2.3, this experimental result also agreed with the $S = 0.85R\ln 2$ curve in the exchange-model calculation.

The fifth sample demagnetization was carried out with $S = 0.46R\ln 2$ and yielded no significant pressure change down to zero field as shown in Fig. 4.11. This is a strong

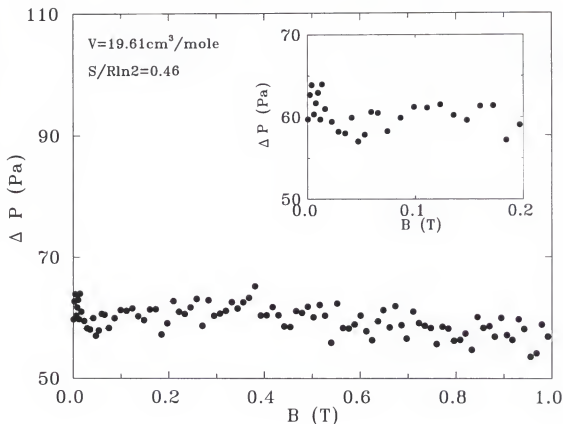


Figure 4.11. Demagnetization data of the $v = 19.61 \text{ cm}^3/\text{mole}$ sample with entropy of $0.46R\ln 2$.

indication that the sample was partially ordered since the pressure change was between those of the 0 and $0.75R\ln 2$ results.

The sixth sample demagnetization had an initial entropy of $0.23R\ln 2$ but was ruined by a large heat leak of approximately 100 nW to the copper bundle. Despite an attempt to cool the bundle by demagnetization, the bundle warmed up from 1 mK to about 5 mK during the 34 hour sample demagnetization. Two probable causes for the large heat leak were the power-supply-ramp unit used in the demagnetization of the sample or the defective lock-in amplifier of the melting pressure thermometer. The thermometer was unusually noisy during the precooling of the sample. Figure 4.12 shows the sixth demagnetization

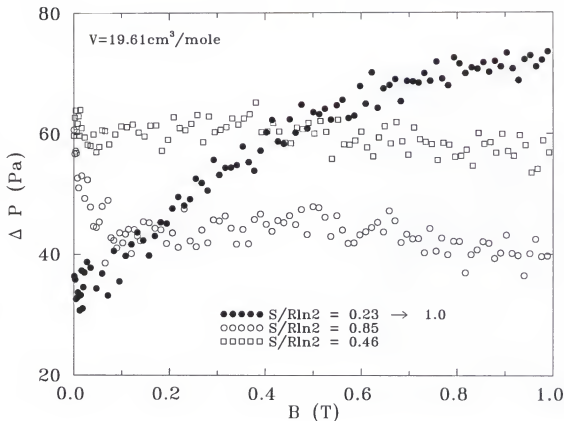


Figure 4.12. Demagnetization data for the $v = 19.61 \text{ cm}^3/\text{mole}$ sample with large heat leak.

curve together with the fourth and fifth demagnetization data. Because of its initial lower entropy, the sample pressure in the sixth demagnetization was higher than those of the fourth and fifth. However, as more heat flowed into the sample, the entropy increased and the pressure dropped with decreasing field. At $B = 0.45 \text{ T}$, the sample entropy had increased to about $0.46R\ln 2$ and to $0.85R\ln 2$ at 0.15 T , becoming almost $R\ln 2$ at zero field.

The seventh sample demagnetization, as shown in Fig. 4.13, was a repetition of the sixth with $S = 0.23R\ln 2$. The overall pressure change during the demagnetization is similar to that of the first demagnetization with $S = 0.02R\ln 2$. In both cases, the pressure measurements show a slight increase from high fields down to approximately 0.2 T , then a

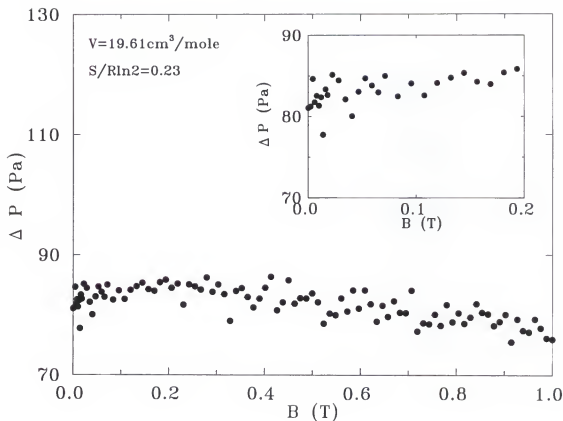


Figure 4.13. Demagnetization data of the $v = 19.61 \text{ cm}^3/\text{mole}$ sample with entropy of $0.23R\ln 2$.

decrease down to zero applied field. The only difference between the two cases was the size of the pressure decrease in low fields. For the seventh sample demagnetization, the total pressure drop was approximately 3 Pa between 0.2 T and 0 T compared to 10 Pa observed in the first demagnetization. As will be presented in Section 4.2.3, this experimental result disagrees with the high-temperature calculations for the multiple-exchange and Heisenberg models.

The demagnetization data clearly show that the pressure of the hcp samples behaves differently depending on the amount of entropy remaining in the sample. High-temperature expansions of the multiple-exchange and Heisenberg models, which are discussed next, give

strong support that pressure increase at low fields of high-entropy samples is the result of adiabatic demagnetization of a disordered ferromagnet. For the low-entropy sample, the best explanation for the pressure decrease at low field is the ferromagnetic ordering transition of the hcp solid.

4.2.3 Comparison of Results with Model Calculations

Two theoretical models, multiple-exchange and Heisenberg, were used in simulation of the sample demagnetizations. The main purpose of the simulation was to predict the pressure change in an hcp sample during a direct demagnetization for comparison with the experimental results. These are high-temperature expansions valid only for $T > T_c$, the ordering temperature.

Exchange Model

The calculation is based on the expression for the free energy provided by Roger *et al.* [23] of their high-temperature expansion of the two-, three-, and four-particle exchange model (for more detail see Section 1.2.2). The expansion, for a sample located in an arbitrary field, was carried to the fourth order of $1/k_B T$. The expression for the free energy for an hcp solid is given by

$$F = -RT \left[\ln(2 \cosh x) + \frac{1}{k_B T} \left(\frac{1}{2} k_B \theta_w p^2 + \frac{3}{4} K'_{sq} p^4 \right) + (1 - p^2) \sum_{n=2}^4 \frac{a_n(p^2)}{2^n n!} \left(\frac{1}{k_B T} \right)^n \right], \quad (4.3)$$

where R is the gas constant; x is a dimensionless parameter given by $\gamma \hbar B_0 / 2k_B T$, with B_0 the applied field; $p (= \tanh x)$ is the polarization; $k_B \theta_w = 3\bar{J}_1 + 3\bar{J}_2 + 9K'_{sq}$ with $\bar{J}_1$ and $\bar{J}_2$ the in-plane nearest-neighbor and out-of-plane nearest-neighbor contributions of the two-particle and three-particle exchanges, respectively, and K'_{sq} the four-particle out-of-basal

plane exchange; and the $a_n(p^2)$ are polynomials of order $2n - 1$ in p^2 whose coefficients are shown in Table 4.8. By taking the derivative with respect to the temperature at constant field and molar volume, $-\partial F/\partial T|_{B_0, v}$, one obtains an expression for the entropy

$$\begin{aligned}
 S = R \bigg\{ & \ln(2 \cosh x) - xp - \frac{x}{k_B T \cosh^2 x} (k_B \theta_w p + 3K'_{sq} p^3) \\
 & + \frac{2xp}{\cosh^2 x} \sum_{n=2}^4 \frac{a_n(p^2)}{2^n n!} \left(\frac{1}{k_B T} \right)^n \\
 & - (1 - p^2) \sum_{n=2}^4 \left[\frac{x}{\cosh^2(x)} \frac{\partial a_n(p^2)}{\partial p} + (n - 1) a_n(p^2) \right] \frac{1}{2^n n! (k_B T)^n} \bigg\}, \quad (4.4)
 \end{aligned}$$

where $\partial a_n(p^2)/\partial x$ is the partial derivative of the polynomials $a_n(p^2)$ with respect to x .

The expression for the pressure can be obtained from the free energy by taking its derivative with respect to volume,

$$P = R \left[\frac{\partial \theta_w p^2}{\partial v} \frac{1}{2} + \frac{\partial K'_{sq}/k_B}{\partial v} \frac{3p^4}{4} + T(1 - p^2) \sum_{n=2}^4 \frac{\partial a_n(p^2)/\partial v}{2^n n!} \left(\frac{1}{k_B T} \right)^n \right], \quad (4.5)$$

where the $\partial a_n(p^2)/\partial v$ is the partial derivative of the polynomials $a_n(p^2)$ with respect to the molar volume v .

A computer program was written to calculate the pressure change during an adiabatic demagnetization of a hcp sample. The program first calculates the entropy for given initial temperature and field. The field was then reduced in steps to simulate the demagnetization. At each new value of the field a root-finding routine, "Newton-Raphson and Bisection" [48], was applied to Eq. 4.4 with the entropy held constant, in order to determine the new temperature T . This new value was substituted into Eq. 4.5 to calculate the pressure change. Figure 4.14 shows the results of the calculation at different entropies for an hcp sample of molar volume of $19.65 \text{ cm}^3/\text{mole}$. Since the model breaks down at low temperatures (fields),

Table 4.8. Polynomial coefficients, obtained from Ref. [23], for $A_{\alpha\beta\gamma}(p^2)$ of the exchange model for $n = 2$ to 4. $A_{\alpha\beta\gamma}(p^2)$ are related to the polynomial $a_n(p^2)$ of the high-temperature expansion by $a_n(p^2) = \sum \tilde{J}_1^\alpha \tilde{J}_2^\beta K^\gamma A_{\alpha\beta\gamma}(p^2)$. For a given n and each combination of $\tilde{J}_1^\alpha \tilde{J}_2^\beta K^\gamma$, the coefficients are listed in columns with increasing power of p^2 up to $(p^2)^{2n-1}$.

n=2	$\tilde{J}_1^2$	$\tilde{J}_2^2$	$\tilde{J}_1\tilde{J}_2$	$\tilde{J}_1K$	$\tilde{J}_2K$	K^2
p^0	9	9	0	18	18	42.75
p^2	33	33	72	228	228	339.75
p^4				66	66	212.25
p^6						29.25

n=3	$\tilde{J}_1^3$	$\tilde{J}_2^3$	$\tilde{J}_1^2\tilde{J}_2$	$\tilde{J}_1\tilde{J}_2^2$	$\tilde{J}_1^2K$
p^0	18	-18	0	108	135
p^2	222	246	1512	1440	4257
p^4	-864	-876	-3024	-2988	-7479
p^6					-3681

	$\tilde{J}_2^2K$	$\tilde{J}_1\tilde{J}_2K$	$\tilde{J}_1K^2$	$\tilde{J}_2K^2$	K^3
p^0	135	432	405	405	308.35
p^2	4257	10872	15849	15849	14880.75
p^4	-7479	-15408	-15363	-15363	-5431.50
p^6	-3681	-7704	-22005	-22005	-29344.50
p^8			-4230	-4230	-12744.75
p^{10}					-1460.25

n=4	$\tilde{J}_1^4$	$\tilde{J}_2^4$	$\tilde{J}_1^3\tilde{J}_2$	$\tilde{J}_1\tilde{J}_2^3$	$\tilde{J}_1^2\tilde{J}_2^2$
p^0	-306	-162	0	-288	936
p^2	1494	3798	30528	25920	55800
p^4	-28458	-31194	-182016	-176544	-302904
p^6	36846	38286	181440	178560	285624

	$\tilde{J}_1^3K$	$\tilde{J}_2^3K$	$\tilde{J}_1^2\tilde{J}_2K$	$\tilde{J}_1\tilde{J}_2^2K$	$\tilde{J}_1^2K^2$
p^0	-576	-288	5184	4320	3744
p^2	75528	82536	389136	368112	566304
p^4	-484056	-489912	-1798752	-1781184	-2300040
p^6	284184	286200	887760	881712	-132144
p^8	261432	262584	854208	850752	2067048
p^{10}					542160

	$\tilde{J}_2^2K^2$	$\tilde{J}_1\tilde{J}_2K^2$	$\tilde{J}_1K^3$	$\tilde{J}_2K^3$	K^4
p^0	3744	19440	11088	11088	-1227.375
p^2	566664	1338912	1323036	1323756	951497.625
p^4	-2301360	-4959744	-3793392	-3796080	-1763533.875
p^6	-130320	-447456	-3547176	-3543840	-4674730.875
p^8	2065968	4252752	4394976	4393896	2087095.875
p^{10}	542376	1118592	3051468	3050964	3891274.875
p^{12}			437760	437976	1265529.375
p^{14}					121854.375

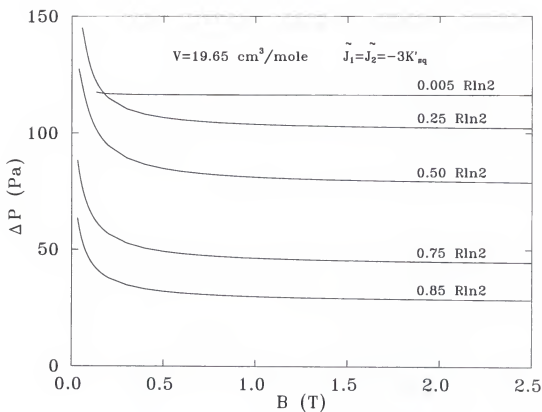


Figure 4.14. Calculated pressure change relative to the unpolarized state using the high-temperature expansion of the multiple-exchange model during a sample demagnetization at different initial entropies.

the calculation in this range is excluded from the graph. Except for only a small pressure increase for the lowest entropy, all curves show large upward turns of the pressure at low fields. Our experimental data with $S = 0.72R\ln 2$ and $0.85R\ln 2$ agreed qualitatively with the calculation; however, the pressure of a lower-entropy samples did not behave according to the theoretical prediction. We interpret this to mean that the condition $T > T_c$ required for validity of the expansion is not met because T_c has been reached.

In the simulation we chose $\tilde{J}_1 = \tilde{J}_2$ as assumed by Roger *et al.* [23] and then used the precooling pressure data of the Tokyo group [6] to determine the value of K'_{sq} . For their hcp sample of $v = 19.66 \text{ cm}^3/\text{mole}$, they found $\partial\theta_w/\partial v = 33.6 \text{ } \mu\text{K mole}/\text{cm}^3$ and $1/k_B(\partial K'_{sq}/\partial v) = -3.8 \text{ } \mu\text{K mole}/\text{cm}^3$. Since $k_B\theta_w = 3\tilde{J}_1 + 3\tilde{J}_2 + 9K'_{sq}$, we have

$$\begin{aligned} \frac{\partial\theta_w}{\partial v} &= \frac{1}{k_B} \left(6\frac{\partial\tilde{J}_1}{\partial v} + 9\frac{\partial K'_{sq}}{\partial v} \right) = \frac{6}{k_B} \frac{\partial\tilde{J}_1}{\partial v} + 9(-3.8) = 33.6 \text{ } \mu\text{K mole}/\text{cm}^3 \quad (4.6) \\ \Rightarrow \frac{1}{k_B} \frac{\partial\tilde{J}_1}{\partial v} &= 11.3 \text{ } \mu\text{K mole}/\text{cm}^3 \approx -\frac{3}{k_B} \frac{\partial K'_{sq}}{\partial v}. \end{aligned}$$

The above result indicates that the value for K'_{sq} from the pressure measurement is approximately 5/3 times larger than the estimation for K'_{sq} by Roger *et al.* [23] who used the susceptibility data at $v = 19.4 \text{ cm}^3/\text{mole}$ [2]. Other susceptibility data [49] and calculations³ yields $\tilde{J}_1 = -3.91K'_{sq}$ and $-4.82K'_{sq}$ respectively for $v = 19.4 \text{ cm}^3/\text{mole}$. We believe that the pressure data yields a better estimation since they were obtained at high temperature (during the precooling of the sample) where the expansion is valid. The use of the susceptibility data for estimation of the exchange parameters is less preferable because one is forced to make the fitting at a very low temperature where the high-temperature expansion may not be valid.

³D. Ceperley, private communication.

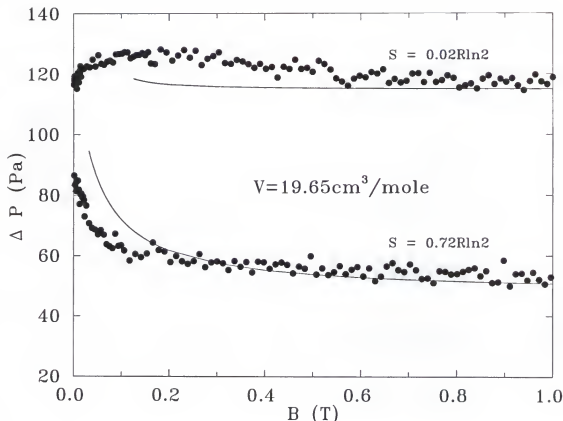


Figure 4.15. Pressure change (solid dots) relative to the unpolarized state during a sample demagnetization at different initial entropies and calculation from the high-temperature expansion of the multiple-exchange model (lines) for $v = 19.65 \text{ cm}^3/\text{mole}$.

Comparing this experimental result with that for the $S = 0.72R\ln 2$ high-temperature expansion in Fig. 4.15, we find that both of them have increasing pressures for decreasing field at low fields. The magnitude of the pressure rise and the field value at which this rise occurs are also similar. These calculated results provide strong evidence that the observed pressure increase is the result of adiabatic demagnetization of a disordered ferromagnet. For the low-entropy sample with $S = 0.02R\ln 2$, the multiple-exchange model also predicts a small pressure increase; however, that was not observed for that case. The propable explanation is that the decrease in pressure is because of the ferromagnetic ordering signature of the hcp solid ^3He . The high-temperature series expansion would not be valid there.

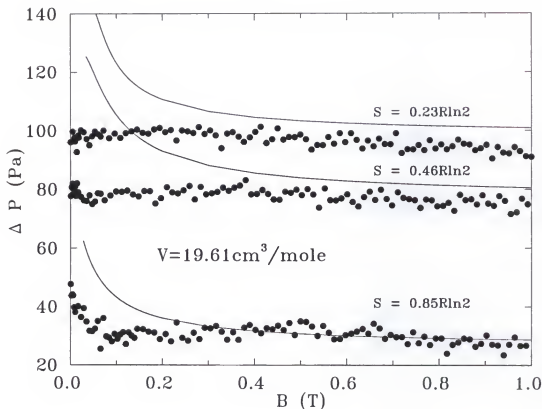


Figure 4.16. Pressure change (solid dots) during a sample demagnetization at different initial entropies and calculations from the high-temperature expansion of the exchange model (lines) for $v = 19.61 \text{ cm}^3/\text{mole}$.

Figure 4.16 shows the plot of the calculated pressure together with experimental data for the second hcp sample $v = 19.61 \text{ cm}^3/\text{mole}$. The data for $S = 0.85R\ln 2$ exhibited an increase in pressure at low fields, qualitatively in agreement with the calculated result although not quite as large. The disagreement in the pressure magnitude may be due to the fact that the high-temperature expansion of the model has been carried only to the fourth order of $1/k_B T$ or because of incorrect estimation in the values of the three exchange parameters $\tilde{J}_1$, $\tilde{J}_2$, and K'_{sq} . Our simulation using the high-temperature expansion up to 10th order of $1/k_B T$ of the Heisenberg model (discussed in the next section) yielded a

smaller change in pressure at low fields than that obtained from the multiple-exchange model for the same entropy value.

For the lower-entropy data of $S = 0.46$ and $0.23R\ln 2$, there was large disagreement with the calculated results. The $S = 0.46R\ln 2$ data had only a small pressure increase of approximately 5 Pa at low fields; however, the calculated pressure change for the same entropy yielded a positive change in ΔP of more than 40 Pa. For $S = 0.23R\ln 2$, the experimental data had a 3 Pa pressure decrease near zero field while the exchange model predicted a positive pressure change of more than 40 Pa. These large disagreements indicated that there was magnetic ordering of the nuclear spins.

Because of large uncertainty in the pressure during the sample precoolings (Figs. 4.6 and 4.7), the experimental data have been shifted so that they agree with the calculation at high fields where the calculated pressure change is insignificant. In both figures, the measurements of high-entropy samples qualitatively agree with the calculated results. For lower-entropy samples, the calculation fails to predict the pressure change at low fields. The discrepancy becomes larger with decreasing entropy, giving strong evidence that nuclear spins in the sample are more strongly ordered at low entropy.

Heisenberg One-Exchange Model

To check for the validity of our multiple-exchange model simulation and to extend our calculation to lower temperature (field), we employed the high-temperature expansion of the Heisenberg model up to the tenth order in $J/k_B T$ where J is the interaction of two

nearest neighbors [50]. The free energy for this model is given by⁴

$$F = -RT \left[\ln(2 \cosh x) + \sum_{n \geq 1} \sum_{0 \leq j \leq n} \frac{y^n}{2^n n!} f(n, j) p^{2j} \right], \quad (4.7)$$

where $x = \hbar\gamma B_0/2k_B T$ and $p = \tanh x$ are the same as in Eq. 4.3 and $y = J/k_B T$. The high-temperature coefficients, $f(n, j)$, are listed in Table 4.9 for values of $n \leq 8$ and the coefficients $f(9, 0)$, $f(9, 1)$, $f(10, 0)$, and $f(10, 1)$ included for completeness. These values were calculated for a face-centered-cubic (fcc) solid; however, they can be used for an hcp solid because both solids have 12 nearest neighbors.⁵

The entropy, obtained by taking the temperature derivative of the free energy, is given by

$$S = R \left[\ln(2 \cosh x) - xp + \sum_{n \geq 1} \sum_{0 \leq j \leq n} \frac{y^n}{2^n n!} f(n, j) p^{2j} \left(1 - n - \frac{2jx}{p \cosh^2 x} \right) \right]. \quad (4.8)$$

The expression for the pressure can be determined by taking the derivative of the free energy with respect to the volume, giving

$$P = N_A \frac{\partial J}{\partial v} \sum_{n \geq 1} \sum_{0 \leq j \leq n} \frac{y^{n-1}}{2^n (n-1)!} f(n, j) p^{2j}. \quad (4.9)$$

It is easy to show that the multiple-exchange model with $\bar{J}_1 = \bar{J}_2 = J$ and $K'_{sq} = 0$ is equivalent to the Heisenberg model. For example, the first order term of Eq. 4.3 can be written as

$$F = -\frac{R\theta_w}{2} p^2 = \frac{-3RJ}{k_B} p^2. \quad (4.10)$$

⁴Note that the first term in the free energy is just the Zeeman energy term which is also contained in the free energy for the multiple-exchange model.

⁵With the assumption that the in-plane and out-of-basal plane interactions are the same ($\bar{J}_1 = \bar{J}_2$) and the four-particle exchange process is negligible ($K'_{sq} = 0$) in the hcp solid.

Table 4.9. High-temperature expansion coefficients, obtained from Ref. [50], of the Heisenberg model. For a given $n \leq 8$, $f(n, j)$ are listed with increasing $0 \leq j \leq n$. Coefficients for $n = 9, 10$ are not completely determined; however, available values of $f(9, 0)$, $f(9, 1)$, and $f(10, 0)$ are included.

n	f.c.c lattice			
1	0	6		
2	18	120	-138	
3	108	3312	-11172	7752
4	180 -720756	117360	-838656	1441872
5	-5040 -22164080	5104416 94744224	-66190560	208511040
6	162000 -57308461200	263405088 50330952000	-5631588672 -16184350080	28529880864
7	14565600 -12014728917408	15717292800 17682684163200	-518612953152 -12492230849280	3916735549440 3410421148800
8	563253408 -2409974307485952 -855895279911840	1063892512512 5261160226823424	-51595772643456 -6105026127070080	552101473737984 3608165330784000
9	17544639744	80532234584064		
10	750651187968			

If one evaluates only the $n = 1$ terms in Eq. 4.7, one gets

$$F = \frac{-RTy}{2} (f(1, 0) + f(1, 1)p^2) = \frac{-3RJ}{k_B} p^2, \quad (4.11)$$

which is identical to that of the exchange model. For $n = 2$ the free energy for the multiple-exchange model has the form:

$$\begin{aligned}
 F &= -RT(1 - p^2) \frac{a_2(p^2)}{8(k_B T)^2} \\
 &= -N(1 - p^2) \frac{9\tilde{J}_1^2 + 9\tilde{J}_2^2 + (33\tilde{J}_1^2 + 33\tilde{J}_2^2 + 72\tilde{J}_1\tilde{J}_2)p^2}{8(k_B T)} \\
 &= -N(1 - p^2) \frac{18J^2 + 138J^2p^2}{8(k_B T)^2} = -N \frac{J^2}{8(k_B T)} (18 + 120p^2 - 138p^4).
 \end{aligned} \quad (4.12)$$

The free energy of the Heisenberg nearest-neighbor model yields

$$\begin{aligned}
 F &= -RT \frac{y^2}{8} [f(2,0) + f(2,1)p^2 + f(2,2)p^4] \\
 &= -N \frac{J^2}{8(k_B T)} (18 + 120p^2 - 138p^4).
 \end{aligned} \tag{4.13}$$

Again, the two models give the same result for a single exchange parameter and one can show that up to $n = 4$ the two expansions have the same form providing that $\tilde{J}_1 = \tilde{J}_2 = J$ and $K'_{sq} = 0$ in the multiple-exchange model.

To calculate the pressure change during a sample demagnetization to lower temperature, we employed the high-temperature expansion to the 10th order in $1/k_B T$ of the Heisenberg model. The simulation was carried out in the same manner as described for the multiple-exchange model. The results, shown in Fig. 4.17, indicate a pressure increase at low fields for all entropies. These results validate our simulated data of the multiple-exchange model and confirm our interpretation that the pressure increase at low fields of high-entropy sample is a consequence of adiabatic demagnetization of a disordered ferromagnet.

High-temperature expansion of the multiple-exchange and Heisenberg models can only explain the pressure data of high-entropy samples. For both models, it fails to predict the pressure change of a sample with low entropy. This would be expected since the expansion fails for $T \lesssim T_c$. We conclude that the pressure behavior observed for entropies of 0.46, 0.23, and $0.02R \ln 2$ are the results of magnetic ordering in hcp solid ^3He . In this experiment the temperature of the spins cannot be measured during the demagnetization; however, the ordering temperature T_c can be estimated. Based on the susceptibility data [3] and the calculated exchange frequencies shown in Table 4.10 for hcp solid⁶, the T_c of our sample is

⁶D. Ceperley, private communication.

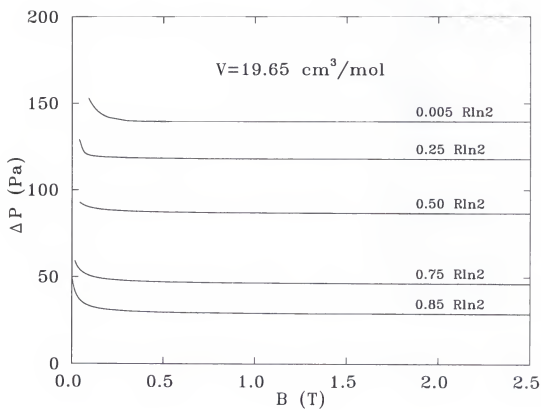


Figure 4.17. Calculated pressure change using the high-temperature expansion of the Heisenberg model during a sample demagnetization at different initial sample entropy.

Table 4.10. Calculated frequencies of exchange cycles in hcp solid ^3He for $v = 19.4 \text{ cm}^3/\text{mole}$ by D. Ceperley (private communication).

Name	Frequency (μK)	Error (%)
J_{nn}	2.4	11
J'_{nn}	3.3	10
J_T	4.8	11
J'_T	2.4	8
J_T^*	0.5	13
K'_{sq}	1.4	15

approximately $17 \mu\text{K}$ (assuming that T_c varies as $v^{18 \pm 1}$). If the relation $B/T = \text{constant}$ for non-interacting spins were followed, the field at $T = 17 \mu\text{K}$ would be $2.5(17/614) = 0.07 \text{ T}$. From Fig 4.8 one can see that for $B \leq 0.07 \text{ T}$ the pressure of the sample drops rapidly with decreasing field indicating a significant change in the magnetic state of the nuclear spins. Thus, we conclude that $T_c = 17 \pm 3 \mu\text{K}$.

CHAPTER 5

SUMMARY AND FUTURE WORK

Theoretical calculations [1] have predicted that the hcp solid will order ferromagnetically with an ordering temperature of $20\text{ }\mu\text{K}$ [23]. Such a low temperature makes the study of the magnetic properties of the hcp solid difficult. Magnetization [2, 3] and pressure [4] measurements of hcp solids for $T > T_c$ have indicated a strong ferromagnetic tendency. However, no magnetic transition was observed because the ordering temperature could not be reached using the indirect cooling method.

To achieve lower temperature, ^3He samples were directly demagnetized, an idea that has been around for many years [5]. Using the sample-demagnetization technique, we carried out pressure measurements for one bcc and two hcp samples. With a better design of the experimental cell, we have reduced heating near the end of the sample demagnetization and eliminated the difficulty encountered by the Tokyo group. For the first time, we were able to observe and identify the signature of magnetic ordering in the hcp solid ^3He . Below are the summaries of our pressure measurements on bcc and hcp solids.

5.1 bcc Solid

Our bcc sample with a molar volume of $20.10\text{ cm}^3/\text{mole}$ was used as a test of our experimental arrangement. The sample was precooled to 0.55 mK in a field of 2.5 T , resulting in the removal of nearly $0.99R\ln 2$ of the entropy. During the sample demagnetization, we observed magnetic transitions occurring when the sample entered the HFP and the LFP. The observation of the LFP transition indicated that our sample was cooled to a temperature

below $30\text{ }\mu\text{K}$. Near zero field, the pressure of the sample remained constant, indicating a much smaller heat flow into our sample than in that of the Tokyo group [6] who observed a pressure drop of approximately 35 Pa between 0.01 and 0 T. This knowledge is important in interpreting the results of our hcp samples.

5.2 hcp Solid

We made pressure measurements on two hcp samples with molar volumes of 19.65 and $19.61\text{ cm}^3/\text{mole}$. There were three demagnetizations for the $19.65\text{ cm}^3/\text{mole}$ sample and each was carried out at different remaining entropies after a precooling. In the first demagnetization with approximately $0.02R\ln 2$ entropy, we found that the pressure decreased by 10 Pa as the field was reduced from 0.2 to 0 T. Both the second and third demagnetizations were with approximately $0.75R\ln 2$ remaining entropy but at two different temperatures of the copper bundle. In both cases, we found a pressure increase in low fields in contrast with the $0.02R\ln 2$ sample. For the second demagnetization, the pressure rose by 26 Pa between 0.1 and 0 T.

The fourth demagnetization was for the $19.61\text{ cm}^3/\text{mole}$ sample with a remaining entropy of $0.85R\ln 2$ and we observed a pressure increase of 20 Pa between 0.1 and 0 T. The fifth demagnetization of $0.46R\ln 2$ entropy yielded zero pressure change down to zero field. The sixth demagnetization with $0.23R\ln 2$ was ruined by a large heat leak and the seventh, a repeat of the sixth, yielded a 3 Pa pressure decrease between fields of 0.2 and 0 T.

Simulated demagnetizations of multiple-exchange and Heisenberg models indicate a pressure rise in low field for all entropies. However, we observed such an increase in pressure only for samples with $0.75R\ln 2$ entropies or higher. Therefore, we conclude that the pressure

decrease observed in the demagnetization of a low-entropy sample is the ferromagnetic ordering signature of the hcp solid.

5.3 Future Work on Solid ^3He

Although our pressure measurements show clear evidence of magnetic ordering in hcp, there are other experiments which could be done to further our understanding of the magnetic properties of hcp solid. Two such experiments are ac magnetic-susceptibility measurement of hcp solid and the pressure measurement of a bcc-hcp mixture, which will not only provide information on the magnetic properties of the hcp solid but also of the bcc solid.¹

In addition, we suggest the measurements of B_{c1} as a function of v using the direct demagnetization method for a better determination of the exchange parameters of the bcc solid. For the hcp solid, we proposed the measurements of $P_v(T, B_0)$ at different v . Below are general description of these experiments.

5.3.1 AC Susceptibility Measurement of hcp ^3He

The goal of this experiment would be to study the magnetic properties of hcp solid in its ordered state. Our pressure measurements of hcp samples show that the direct nuclear demagnetization method can cool a sample into its ordered state. This cooling method therefore would be used in the susceptibility experiment. During the cooling and warming processes, the magnetic susceptibility of the sample would be measured so that the magnetic state of the sample could be studied both above and below the transition.

Although a new experimental cell would be needed for the susceptibility measurement, the same 2.5 T magnet used in the pressure measurements can be used to provide the

¹E. D. Adams, P. Kumar, and Y. Takano, private communication.

precooling field for the sample. The experimental cell, made from pure silver, would have two identical cylindrical cavities packed with silver sinter. One cavity would be used as the ^3He cell and the other, kept in vacuum, would be used as a dummy for cancelling out the background of the silver cell. The pick-up coil, consisting of an astatic pair would be wound directly onto the body of the cell, and the secondary, primary, and compensation coils would be wound on a former situated inside the static-field coil.

In the experiment, an hcp sample with density near the bcc-hcp line would be formed² and then directly demagnetized to the ordered state with precooling and demagnetization processes similar to those described in Sections 4.2.1 and 4.2.2. The field dependence of the magnetic susceptibility would then be measured during the cooling and warming of the sample. The susceptibility behaviors above and below the ordering temperature should yield useful information about the ordered state of hcp solid ^3He .

5.3.2 Pressure Measurement of a bcc-hcp Mixture

The bcc-hcp phase line is essentially flat in the $P - T$ plane for temperatures near 1 K, and solid formed with a molar volume of about $19.8 \text{ cm}^3/\text{mole}$ would have a mixture of bcc and hcp structures [45]. However, in the vicinity of T_N and T_c , the slope dP/dT may be large. Pressure measurement of the mixture would map out the phase line, the slope of which would yield important information on the magnetic characteristics of the mixture.

If either the bcc or the hcp solid becomes magnetically ordered, there will be a large change in the pressure slope because of a large reduction in the entropy of the ordered

²To have the highest ordering temperature.

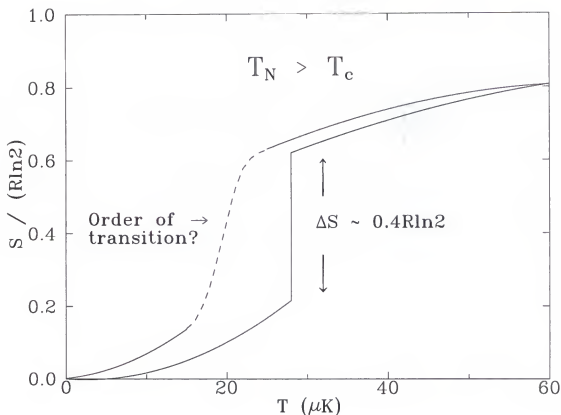


Figure 5.1. Entropy curves of the bcc-hcp mixture for $T_N > T_c$.

solid. The possible magnitude of this pressure change can be estimated using the Clausius-Clapeyron equation

$$\frac{dP}{dT} = \frac{S_{hcp} - S_{bcc}}{v_{hcp} - v_{bcc}}, \quad (5.1)$$

where S_{hcp} and S_{bcc} are the entropy values of the hcp and the bcc solids respectively and v_{hcp} and v_{bcc} are the molar volumes of the solids. The difference $v_{hcp} - v_{bcc}$ is approximately $-0.1 \text{ cm}^3/\text{mole}$ at 1 K [45] and decreases somewhat as the temperature decreases.

If the bcc solid ordered first (*i.e.* $T_N > T_c$), there would be a reduction of approximately $0.4R \ln 2$ in the entropy of the bcc solid [29]. Assuming that the entropy of the unordered hcp solid has the same value as that of the bcc solid above T_N , and inserting these values

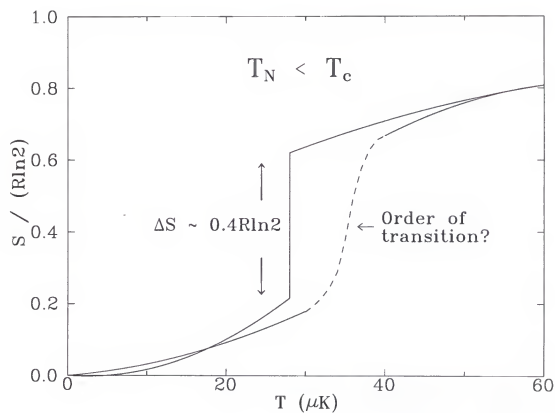


Figure 5.2. Entropy curves of the bcc-hcp mixture for $T_N < T_c$.

into Eq. 5.1, one obtains

$$\frac{dP}{dT} \approx \frac{0.4R \ln 2}{-0.1 \text{ cm}^3/\text{mol}} \approx -20 \text{ Pa}/\mu\text{K}. \quad (5.2)$$

However, if the hcp solid orders first ($T_c > T_N$) then

$$\frac{dP}{dT} \approx +20 \text{ Pa}/\mu\text{K}. \quad (5.3)$$

Figures 5.1 and 5.2 show the entropy curves for the two cases. In either case, the slope change should be close to $20 \text{ Pa}/\mu\text{K}$ which is a magnitude larger than that of the melting curve near 1 mK. Such a large pressure slope could be easily detected by our current experimental cell.

The mixture could be cooled to low temperatures by the direct demagnetization method similar to that described in Chapter 3. The pressure change versus time, $P(t)$, of the mixture can be monitored during and after the demagnetization. During the warm up at zero field, the $P(t)$ curve should have a negative slope if the bcc solid has a higher ordering temperature, and a positive slope if the ordering temperature in the hcp solid is higher. Figure 5.3 and 5.4 show the predicted warm-up pressure curves for both cases. The ordering temperature of the mixture at zero field can be estimated from known ordering temperatures at higher molar volumes using the simple power law described in Chapter 1.

5.3.3 Measurement of $B_{c1}(v)$ of bcc ^3He

Our bcc demagnetization data (see Section 4.1.3) indicated a scaling factor of 16.7 for $B_{c1}(v)$ while results from other works [12, 6] yielded the values of 14.5 and 14.7, respectively.

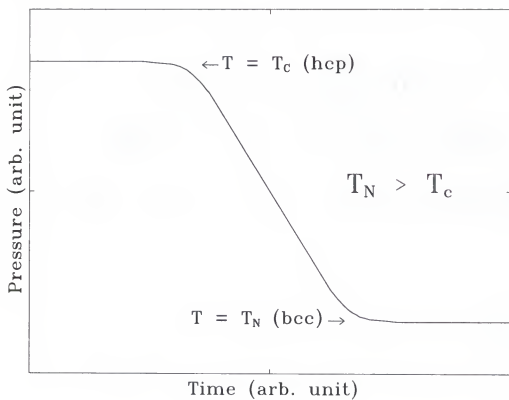


Figure 5.3. The predicted pressure change during a warm up of the bcc-hcp mixture for $T_N > T_c$.

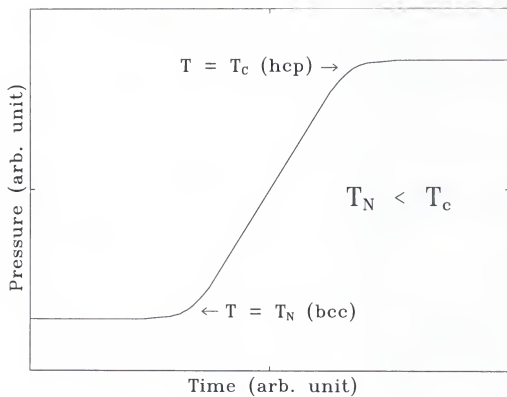


Figure 5.4. The predicted pressure change during a warm up of the bcc-hcp mixture for $T_N < T_c$.

This discrepancy requires further investigation because of its implication for the volume-dependence of various exchange parameters.

To achieve a wider range of molar volumes than that of the previous work [12], the samples could be cooled using the direct demagnetization method. A temperature of less than $30\text{ }\mu\text{K}$ can be achieved with this cooling method allowing access to the LFP phase even for the highest-density bcc solid. Because B_{c1} has a T^4 dependence [13], most of the sample entropy must be removed before the sample demagnetization in order for the sample to enter the HFP-LFP phase line at the lowest temperature where the temperature-dependence of B_{c1} is small, see Fig 4.5. The magnetic state of the sample could be monitored by the pressure change in the sample. A jump in pressure during the demagnetization as shown in Fig. 4.4 indicates that the sample has crossed the HFP-LFP boundary.

5.3.4 Measurement of Exchange Parameters of hcp ^3He

Because the objective in this work was to cool an hcp sample into its ordered state, we did not attempt to measure the volume-dependence of the exchange parameters $\partial\theta_w/\partial v$ and $\partial K'_{sq}/\partial v$ during the sample precooling with high accuracy. Although, our hcp precooling data are in agreement with those of the Tokyo group [6], there is sizable scatter in the data as shown in Figs. 4.6 and 4.7.

To achieve higher-quality data, we suggest a few modifications during the precooling of the sample. First, the waiting time, at a fixed T_{bundle} , should be longer allowing the temperature of the sample to be close to that of the copper bundle. For example, in Table 4.2, the waiting time at $T_{bundle} = 1.78\text{ mK}$ was only 22 hours causing a large uncertainty in ΔP ; however, if the waiting time were 72 hours or so, the error in ΔP would have been smaller. Second, the excitation voltage of the capacitance bridge should be increased from

2.12 V_{pp} to approximately 10 V_{pp} allowing the sensitivity of the strain gauge to improve from 5 Pa to 2 Pa. Finally, the temperature-dependence of the cell could be reduced further by having a reference capacitor made of the same material (coin-silver) and dimension of the strain gauge and attached directly onto the experimental cell.³

5.4 Conclusions

Direct nuclear demagnetization was used to cool high-density solid ^3He into the magnetically ordered phases. The entropy of the samples was removed by precooling to low temperatures in a field of 2.5 T. The pressure changes in the samples during precooling and demagnetization were detected by a capacitive strain gauge. Two magnetic transitions were observed in a bcc sample were observed with a remaining entropy of approximately $0.01R\ln 2$. Pressure data of two hcp samples at various entropies indicated different behavior near zero field. Results from the high-temperature expansion of the Heisenberg model can explain only the pressure data with remaining entropies of $0.72R\ln 2$ or higher. We conclude that the pressure anomalies observed in the samples with entropies of 0.46, 0.23 and $0.02R\ln 2$ were the results of magnetic ordering in hcp solid ^3He .

For further study of magnetism in solid ^3He , we suggested two future experiments: the ac susceptibility measurements of a hcp solid and pressure measurements of a bcc-hcp mixture. Additional measurements of $B_{c1}(v)$ of bcc solid, and exchange parameters in hcp solids could also be made using current experimental setup. These experiments would yield valuable information about the magnetic properties of solid ^3He .

³Then both the strain gauge and the reference capacitor would have the same temperature and field.

REFERENCES

- [1] M. Roger, J. H. Hetherington, and J. M. Delrieu. *Rev. Mod. Phys.*, 55:1, 1983.
- [2] Y. Takano, N. Nishida, Y. Miura, H. Fukuyama, H. Ishimoto, and S. Ogawa. *Phys. Rev. Lett.*, 55:1490, 1985.
- [3] H. Yano, H. Kondo, T. Suzuki, Y. Minamide, T. Kato, Y. Miura, and T. Mamiya. *Phys. Rev. Lett.*, 65:3401, 1990.
- [4] Y. Miura, S. Abe, S. Sugiyama, T. Mamiya, and R. C. Richardson. In G. G. Ihas and Y. Takano, editors, *Quantum Fluids and Solids*, page 267. AIP, New York, 1989.
- [5] See for example A. Abragam. *C. R. Acad. Sc. Paris*, 284:255, 1977.
- [6] T. Okamoto, H. Fukuyama, H. Akimoto, H. Ishimoto, and S. Ogawa. *Phys. Rev. Lett*, 72:868, 1994.
- [7] S. Abe, T. Takeuchi, H. Kondo, T. Suzuki, H. Yano, Y. Miura, and T. Mamiya. *Phys. Rev. Lett.*, 70:473, 1993.
- [8] R. A. Guyer, R. C. Richardson, and L. I. Zane. *Rev. Mod. Phys.*, 43:532, 1971.
- [9] W. Ni, J. S. Xia, E. D. Adams, P. S. Haskins, and J. E. McKisson. *J. Low Temp. Phys*, 99:167, 1995.
- [10] F. D. C. Willard. *La Recherche*, 11:972, 1980.
- [11] T. Hata, S. Yamasaki, T. Kodama, and T. Shigi. *J. Low Temp. Phys*, 71:193, 1988.
- [12] H. Fukuyama, T. Okamoto, T. Fukuda, H. Akimoto, and S. Ogawa. *Phys. Rev. Lett.*, 67:1274, 1991.
- [13] J. S. Xia, W. Ni, and E. D. Adams. *Phys. Rev. Lett.*, 70:1481, 1993.
- [14] D. D. Osheroff, M. C. Cross, and D. S. Fisher. *Phys. Rev. Lett.*, 44:792, 1980.
- [15] D. D. Osheroff, H. Godfrin, and R. Ruel. *Phys. Rev. Lett.*, 58:2458, 1987.
- [16] D. J. Thouless. *Proc. Phys. Soc.*, 86:893, 1965.
- [17] A. Benoit, J. Bossy, J. Flouquet, and J. Schweizer. *J. Phys. Lett. (Paris)*, 46:L923, 1985.
- [18] W. P. Halperin, C. N. Archie, F. B. Rasmussen, R. A. Buhrman, and R. C. Richardson. *Phys. Rev. Lett.*, 32:927, 1974.

- [19] A. K. McMahan and J. W. Wilkins. *Phys. Rev. Lett.*, 35:376, 1975.
- [20] H. Godfrin and D. D. Osheroff. *Phys. Rev. B*, 38:4492, 1988.
- [21] D. D. Osheroff. *J. Low Temp. Phys.*, 87:297, 1992.
- [22] A. K. McMahan and R. A. Guyer. *Phys. Rev. A*, 7:1105, 1973.
- [23] M. Roger, E. Suaudeau, and M. E. R. Bernier. *Phys. Rev. B*, 35:2091, 1987.
- [24] M. F. Panczyk and E. D. Adams. *Phys. Rev. A*, 1:1356, 1970.
- [25] D. D. Osheroff and C. Yu. *Phys. Lett. A*, 77:458, 1980.
- [26] D. M. Ceperley and G. Jacucci. *Phys. Rev. Lett.*, 58:1648, 1987.
- [27] T. Okamoto, PhD thesis, University of Tokyo, 1991 (unpublished).
- [28] W. P. Kirk and E. D. Adams. *Phys. Rev. Lett.*, 27:392, 1971.
- [29] W. Ni, J. S. Xia, and E. D. Adams. *Phys. Rev. B*, 50:336, 1994.
- [30] R. B. Kummer, R. M. Mueller, and E. D. Adams. *J. Low Temp. Phys.*, 27:319, 1977.
- [31] D. D. Osheroff. *Physica B*, 109:1461, 1982.
- [32] A. Sawada, H. Yano, M. Kato, K. Iwahashi, and Y. Masuda. *Phys. Rev. Lett.*, 56:1587, 1986.
- [33] T. Okamoto, H. Fukuyama, T. Fukuda, H. Ishimoto, and S. Ogawa. *Physica B*, 165:825, 1990.
- [34] E. D. Adams, Y. H. Tang, and K. Uhlig. *J. Low Temp. Phys.*, 84:109, 1991.
- [35] D. S. Greywall and P. A. Busch. *Phys. Rev. B*, 36:6853, 1987.
- [36] E. D. Adams. *Rev. Sci. Instrum.*, 64:601, 1993.
- [37] U. E. Isareleson and C. M. Gould. *Rev. Sci. Instrum.*, 55:1143, 1984.
- [38] E. D. Adams and J. M. Goodkind. *Cryogenics*, 3:83, 1963.
- [39] G. J. Labbe, H. Royce, Y. Takano, and G. G. Ihas. In G.G. Ihas and Y. Takano, editors, *Quantum Fluids and Solids*, page 395. AIP, New York, 1989.
- [40] J. Xu, O. Avenel, J. S. Xia, M-F Xu, T. Lang, P. L. Moyland, W. Ni, E. D. Adams, G. G. Ihas, M. W. Meisel, N. S. Sullivan, and Y. Takano. *J. Low Temp. Phys.*, 89:719, 1992.
- [41] Y. Takano. *Rev. Sci. Instrum.*, 65:1667, 1994.
- [42] R. M. Mueller, C. Buchai, T. Oversluizen, and F. Pobell. *Rev. Sci. Instrum.*, 49:515, 1978.
- [43] M. W. Meisel, G. R. Steward, and E. D. Adams. *Cryogenics*, 29:1168, 1989.

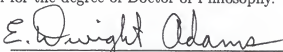
- [44] G. K. White. *Experimental Techniques in Low-Temperature Physics*, page 308. Oxford University Press, Oxford, U. K., 1979.
- [45] G. C. Straty and E. D. Adams. *Phys. Rev.*, 150:123, 1966.
- [46] H. Fukuyama, T. Okamoto, T. Fukuda, H. Akimoto, and S. Ogawa. *Physica B*, 169:197, 1991.
- [47] T. Lang, P. L. Moyland, D. A. Sergatskov, J. Xu, E. D. Adams, and Y. Takano. *J. Low Temp. Phys.*, 101:677, 1995.
- [48] J. C. Sprott. *Numerical Recipes Routines and Examples in BASIC*, page 188. Cambridge University Press, Cambridge, U. K., 1991.
- [49] T. Mamiya, H. Yano, H. Kondo, T. Suzuki, T. Kato, Y. Minamide, Y. Miura, and S. Inoue. *Physica B*, 165:837, 1990.
- [50] G. S. Rushbrooke, Jr. G. A. Baker, and P. J. Wood. Heisenberg model. In C. Domb and M. S. Green, editors, *Phase Transition and Critical Phenomena*, page 245. Academic Press, London, U. K., 1974.

BIOGRAPHICAL SKETCH

Tien Vu Lang was born on May 23rd, 1966, in Saigon, South Vietnam, a son of Anthony Tuyen Lang and Diep Thi Vu.

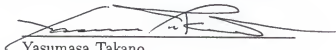
In 1980, he came to the United States as a refugee after escaping from Vietnam. In 1989, he received a B.S. degree in physics and a minor in mathematics from Loyola University in New Orleans. In the same year, he was accepted in the Physics Department graduate program at the University of Florida. He moved to Gainesville, FL, and has worked in the Microkelvin Laboratory since May of 1989. In 1993, he became a Ph.D. candidate and plans to graduate in May of 1996.

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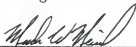
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